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PART VI, Cont.

PROCESS CONTROL

CHAPTER XIV

SAMPLING AND ANALYTICAL METHODS

 $\mathbf{B}\mathbf{y}$

A. H. Case

Separations Chemistry Laboratory

PART VI: PROCESS CONTROL, Cont.

CHAPTER XIV

SAMPLING AND ANALYTICAL METHODS

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PART VI: PROCESS CONTROL, CONT.

CHAPTER XIV

SAMPLING AND ANALYTICAL METHODS

Information required for control of the operation of B Plant processes will be furnished by in-line instrumentation and by laboratory analysis of samples taken from the various tanks and streams. The variations in composition of feed material and the complexity and variety of the processes put heavy demands on the laboratory for versatility and reliability. Adequate methods and equipment used with painstaking workmanship are necessary to insure that the information supplied by the laboratory is valid and timely. To make the laboratory results meaningful, it is essential that the samples delivered to the laboratory be representative of the tank or stream from which they are taken. The sampling equipment has been designed with this in mind but strict adherence to established sampling procedures is required.

A. SAMPLING

1. Sampling Installation and Equipment

The installed sampling equipment consists of shielded boxes containing the sample risers and the necessary piping to circulate the desired process solution through the risers by air jet. A portion of the sample is taken from the sample riser in a plastic pipet on the end of a shielded "trombone." The pipet, ordinarily containing 0.5 milliliters of sample, is transferred to a lead-lined, stainless steel container called a "doorstop" and delivered to the laboratory. "Trombones" and "doorstops" are prepared for use by the laboratory, including decontamination when necessary. The sampling installation and equipment are shown in prints 43062-25, 43062-26, H-2-32654, H-2-32655, H-2-2337, H-3-1398, H-3-1514, and H-2-32514.

2. Sampling Procedure

Procure a trombone and doorstop from the laboratory and check to be sure the doorstop is equipped with a plastic insert and a neoprene gasket on the plug. Follow these steps in obtaining a sample.

- 1) Slowly turn on the air jet to the sampler and check that the sampler is circulating properly. Sample circulation can normally be confirmed by measuring the fluctuating radiation levels on the front side of the sample port. Do not loosen the riser cap while the sample is circulating.
- 2) Remove the doorstop handle and lift the doorstop plug. Attach the syringe to the trombone.

- 3) After the sampler has circulated for 15 minutes, shut off the air jet valve slowly. Remove the riser cap from the sampler, swinging the cap away from the port. With the syringe attached and holding the trombone at an angle, insert the trombone into the port riser, then lower the trombone until it strikes the stop. The sample pipet is now in the sampling position.
- 4) Slowly draw out the syringe plunger 1.5 2.0 ml. Then raise the trombone one inch and slowly press the syringe back to zero. (This forces out the excess sample and empties the tip of the pipet.) Raise the trombone three to four inches and wait five to ten seconds to allow dripping of excess sample solution before continuing.
- 5) Lift the trombone out of the riser and pass the pipet near the chamber of the survey instrument while transferring the sample from the port to the doorstop. A significant reading on the instrument will indicate that a sample has been obtained.
- 6) If the survey instrument indicates that no sample was obtained, repeat steps 3, 4, and 5.
- 7) After a sample is obtained, replace the sample port cap and remove the syringe from the trombone.
- 8) With the trombone tip positioned in the doorstop, turn the assembly slowly to the right while pressing down slightly to engage the pipet in its hole in the bottom of the doorstop well. When the tip engages, the trombone assembly will not turn easily in either direction.
- 9) Unscrew the pipet by turning the trombone handle counterclockwise until the pipet is disengaged from the trombone shaft.
- 10) Remove the plastic trombone from the handle and discard in the receptacle for hot waste.
- 11) The sample port should be promptly restored to normal condition by making a water flush of the riser and then replacing the riser cap.
- 12) Perform a contamination survey and any clean-up which is found to be necessary.
- 13) Attach a sample tag containing all pertinent information to the doorstop and deliver the sample to the laboratory.

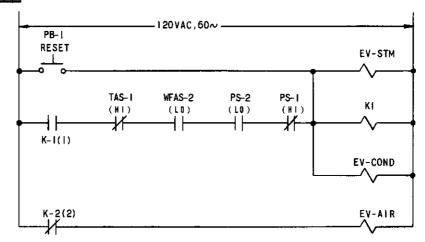


FIGURE XIII-26

Concentrator Safety Interlocks

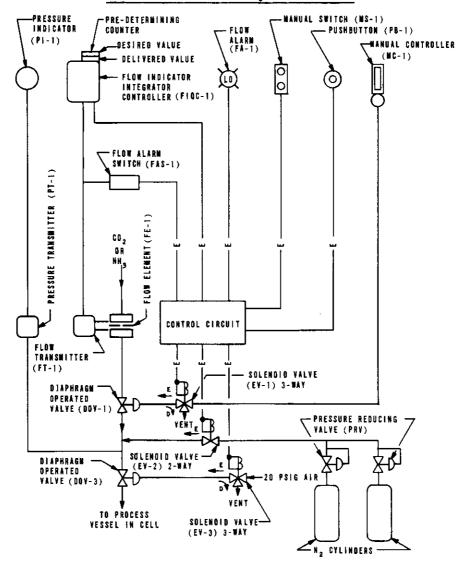


FIGURE XIII-27

Ammonia, Carbon Dioxide and Nitrogen Purge System

Vent System No. 1

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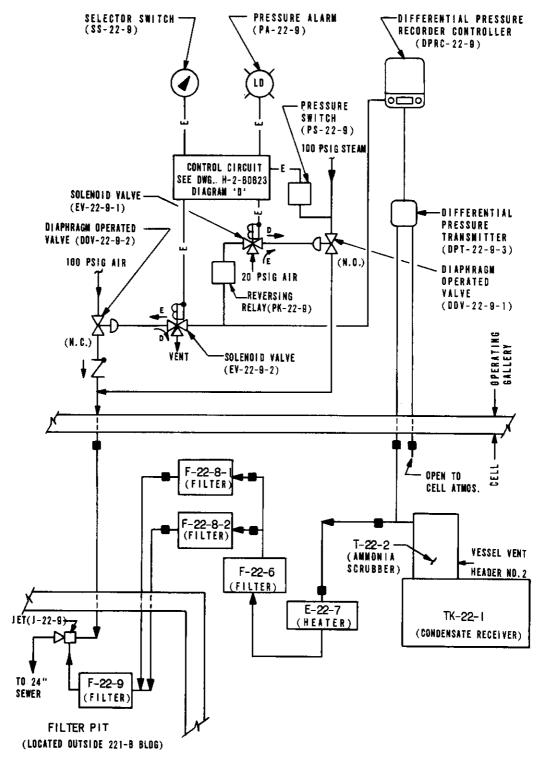
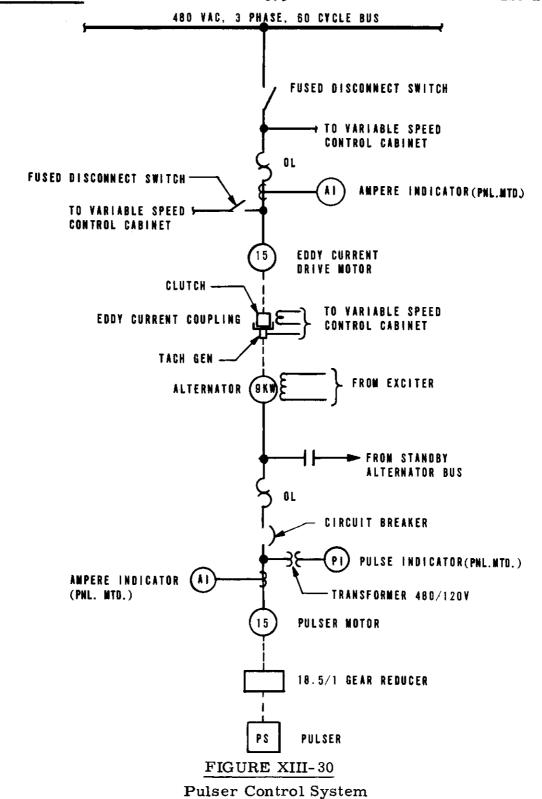


FIGURE XIII-29
Vent System No. 2



This procedure covers the operation of the sampling equipment in use at present in B-Plant. Other types of sampling equipment are planned for the future and procedures for their use will be written as needed.

B. ANALYTICAL METHODS

Following are short descriptions of the general analytical methods used in analyzing B-Plant samples. Where necessary, the methods are preceded by chemical or physical separations to eliminate extraneous materials which would interfere in the analysis.

As plant needs change or as better procedures are developed, other methods may be added or substituted.

1. Counting Methods

Available instrumentation makes it possible to measure the concentration of a radioactive isotope by determining the alpha, beta, or gamma emission rate and using the proper calculations involving specific activity of the isotope, instrument geometry, etc.

- 1) Alpha Alpha emitters must be separated from gross quantities of extraneous solids which could cause low results by absorption. Often two or more alpha emitters can be determined in each others presence by an alpha energy analysis. In some cases a separation is necessary.
- 2) <u>Beta</u> A beta emitter can be determined by counting only after separating from all other beta emitters.
- 3) Gamma In most cases rather complex mixtures of gamma emitters can be analyzed for one or all of the components by a gamma energy analysis. Exceptions would be samples containing isotopes with similar gamma energies or a great disparity in the activity of the isotopes of interest. In these cases a separation is necessary.

Gamma counting can be applied to many non-radioactive isotopes through activation analysis. In this technique, the same is subjected to a controlled exposure in a neutron flux, converting the element of interest to a radioactive species which is determined by gamma energy analysis. Comparison with a standard of known concentration which is given identical treatment makes quantitative results possible.

2. Colorimetric - Spectrophotometric Methods

In many cases the desired constituent of a sample may have a color or can be converted to a compound having a color which is characteristic. The absorbance of the solution for light of a definite

wavelength can be measured on a spectrophotometer and the concentration of the component of interest read from a calibration curve. The wavelength of maximum absorbance may be in the visible, ultraviolet, or infrared portion of the spectrum.

3. Titration Methods

For application of a titrimetric method, the chemical process involved must consist of a single, rapid, stoichiometric reaction between the substance being determined and the standard solution used as a titrant. The point of equivalence is detected by an indicator which shows a marked change in some property (color, potential of an electrode system, etc.) of the titration system. From the concentration of the titrant and the volume required to reach the equivalence point, the concentration of the desired substance may be calculated.

Titrimetric methods may be classified as acid-base titrations, complexometric titrations and oxidation-reduction titrations.

4. Emission Spectrographic Methods

When excited to incandescence the ions of most of the metals and several of the non-metals emit light in wavelength patterns which are characteristic and distinctive for the various ions. In the emission spectrograph the sample is vaporized in an electric arc between two graphite electrodes. The light from the arc is resolved into its various wavelengths by means of a prism or diffraction grating and is focused on a photographic plate. After development, the lines on the plate serve to identify the components of the sample as well as indicate the quantity present.

Although in many cases the results may be considered only semiquantitative, the method is particularly valuable in determining minor impurities in the parts per million range.

5. Flame Photometry and Atomic Absorption

The principle of flame photometry is identical with that of emission spectrography. In this case the sample, in solution, is aspirated into a flame. The intensity of the resultant light of a characteristic wavelength is measured with a photodetector and suitable electronics. When referred to a calibration curve, this intensity indicates the concentration of the elements sought.

Atomic absorption is based on the fact that most of the atoms of an element present in a flame are not heated to incandescence and emit no light. They are, however, in an excited state where they absorb light of the same characteristic wavelength.

In atomic absorption, monochromatic light of the proper wavelength is passed through the flame into which the sample is aspirated. A suitable detector and instrumentation measures the absorption of light and this is converted into concentration by means of a calibration curve.

6. X-Ray Fluorescence

This method may be applied to all except the lightest elements of the periodic table, present in amounts from a few percent to one hundred percent. It is not particularly good for trace amounts.

The primary X-rays from an X-ray tube are allowed to strike the sample, producing a secondary or fluorescent radiation, also in the X-ray portion of the spectrum. This fluorescent radiation with wavelengths characteristic of the emitting elements passes through a collimating slit onto a crystal spectrometer (goniometer) and thence into a detector. The intensity from a given element is affected by the amount of that element present in an almost linear manner.

7. Fluorimetry

Several substances (e.g., uranium) have the property of fluorescence. This is similar to X-ray fluorescence except that the impinging rays are in the ultraviolet and the secondary emission is in the ultraviolet or visible portion of the spectrum. With the proper instrumentation this provides a sensitive method for determining very low concentrations of uranium.

8. Gas Chromatography

Gas chromatography consists of the separation of the components of a mixture of gases or vapors by passage through a column containing a stationary liquid phase on a solid support such as firebrick or celite. The emergence of each component from the column is indicated by a suitable detector and is registered as a peak on a strip chart. By comparison with standards, the components may be identified by the position of the peak (retention time) and quantity may be determined by the area under the peak.

9. Miscellaneous Methods

The following analyses and tests do not fit into any general category but are designed to furnish specific needed information.

9.1 <u>pH</u> - By glass electrode - pH meter. (Electrodes are now available which are sensitive only to certain ions, e.g., Na⁺, Ca⁺⁺, F⁻, Cl⁻, etc. These may find some application in B-Plant control.)

- 9.2 Percent Solids Volume percent undissolved solids is determined visually on a sample allowed to reach equilibrium at a definite temperature and then centrifuged.
- 9.3 <u>Carbonate</u> Potentiometric titration. The carbonate break is determined on an automatic titrator.
- 9.4 Ammonia Ammonia is determined by a Kjeldahl distillation method.
- 9.5 Tests for Tartaric, Citric, and Hydroxyacetic Acids These are qualitative tests to ensure that the correct acid was used in making up process reagents.
- 9.6 Specific Gravity This is defined as the ratio of the weight of a substance to the weight of an equal volume of water. There are several standard methods for measuring specific gravity, e.g., pycnometer and hydrometer.

The following tables show the primary samples and the analyses to be performed. These samples and analyses are based on conceptual flowsheets and operating requirements as seen at the time this chapter was written. Operating experience, altered requirements, and development of improved methods of analysis will no doubt result in additions, deletions, and substitutions in these tables.

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TABLE XIV-1

CENTRIFUGING AND LEACHING FACILITY

Sample	Analysis	Estimated Concentration	Method
PAW	H ⁺	0.5 <u>M</u>	Titration with com-
Tk-11-12	% Solids		plexing Visual estimate after centrifugation
	Sr-90		Chem. Sep. followed by beta counting
	Pm-147		Chem. Sep. followed by beta counting
	Ce-144, ZrNb-95, Ru-103, 106, Sb-125,		Gamma energy analysis
	Ce-137		
	Pu, Np, Am	Low	Chem. Sep. followed by alpha count (and alpha energy analysis if necessary)
	Zr	0.005 <u>M</u>	Colorimetric
Leached PAW Tk-12-1	H ₊	0.5 <u>M</u>	Titration with complexing
	Sr-90		Chem. Sep. followed by beta counting
	Pm-147		Chem. Sep. followed by beta counting
	Ce-144, ZrNb-95, Ru-103, 106, Cs-137		Gamma energy analysis
	Am, Np, Pu	Low	Chem. Sep. followed by alpha count (and alpha energy analy- sis if necessary)
	Fe	0.18 <u>M</u>	Colorimetric
	Na	о.65 <u>м</u>	Flame photometer
	Al	0.05 <u>M</u>	Colorimetric
	U	0.01 <u>M</u>	Chem. Sep.; Fluori- metric determination
	Zr	0.003 <u>M</u>	Colorimetric
	Isotopic Sr (89, 90, inert)		
	Total Metallic Impurities	and ex-	Emission Spectrograph
	Sulfate	0.14 <u>M</u>	
	Nitrate	1.3 <u>M</u>	Colorimetric

TABLE XIV-2

ANALYTICAL REQUIREMENTS FOR PRECIPITATION

Sample	Analysis	Estimated Concentration	Method
PAS Tk-31-3	H ⁺	0.1 <u>M</u>	Titration with complexing
	Sr-90	20 Ci/gal	Chem. Sep. followed by beta counting
	Ce-144		Gamma energy analysis
	Pm-147	10 Ci/gal	Chem. Sep. followed by beta counting
	Am, Np, Pu	Low	Chem. Sep. followed by alpha counting
	Na.	0.4 <u>M</u>	Flame photometry
	Al	0.6 <u>M</u>	Colorimetric
	Fe	0.4 <u>M</u>	Colorimetric
	Ca	0.03 <u>M</u>	Atomic Absorption
	Mg	0.01M	Atomic Absorption
	Mn	0.01 <u>M</u>	Atomic Absorption
	Isotopic Sr (89, 90, inert)		
Supernates and Washes Tk-32-1	рН	ı	Glass electrode-
14-35-1	Sr-90	0.2 Ci/gal	Chem. Sep. followed by beta counting
	Pm-147	0.5 Ci/gal	Chem. Sep. followed by beta counting
	Fission Products		Gamma energy analysis
	Am, Pu, Np	Low	Chem. Sep. followed by alpha count

TABLE XIV-2 (Cont.)

Sample	Analysis	Estimated Concentration	Method
Dissolved	н+	1 <u>M</u>	Titration with complexing
Cake Tk-31-1	Sr-90	200 Ci/gal	Chem. Sep. followed by beta counting
	Pm-147	100 Ci/gal	Chem. Sep. followed by beta counting
	Ce-144		Gamma energy analy- sis
Crude Feed Tk-33-1	н+	1 <u>M</u>	Titration with complexing
	Sr-90	200 Ci/gal	Chem. Sep. followed by beta counting
	Pm-147	100 Ci/gal	Chem. Sep. followed by beta counting
	Am, Np, Pu	low	Chem. Sep. followed by alpha counting
	Fission Products		Gamma energy analysis
	Na	0.1 <u>M</u>	Flame photometry
	Al	0.1 <u>M</u>	Colorimetric
	Fe	0.2 <u>M</u>	Colorimetric
	Ca	0.3 <u>M</u>	Atomic Absorption
	Mg	0.1 <u>M</u>	Atomic Absorption
	Mn	0.01 <u>M</u>	Atomic Absorption
	Pb	0.01 <u>W</u>	Colorimetric or atomic absorption
	U	0.01 <u>W</u>	Fluorimetric after chemical separation
	Isotopic Sr		

TABLE XIV-3

ANALYTICAL REQUIREMENTS FOR SOLVENT EXTRACTION

Sample	Analysis	Estimated Concentration	Method
1 AF Tk-29-2	Нд	4	Glass electrode- meter
	Sr-90		Chem. Sep. followed by beta counting
	Pm-147		Chem. Sep. followed by beta counting
	RuRh-106, ZrNb-95, Ce-144, Cs-137		Gamma energy analysis
	Am, Np, Pu	Low	Chem. Sep. followed by alpha counting
	Na	$1.3 - 1.4 \underline{M}$	Flame Photometry
	Al	0.04-0.1 <u>M</u>	Colorimetric
	Fe	0.1-0.15 <u>M</u>	Colorimetric
	Ca	0.001-0.1 <u>M</u>	Atomic Absorption
	Mg	0.001-0.01 <u>M</u>	Atomic Absorption
	U	0.01-0.02 <u>M</u>	Fluorimetric determination after chem. sep.
	Zr	0.001 <u>M</u>	Colorimetric
	Isotopic Sr		
1AX Tk-28-3	Y-91		Gamma energy analysis
	D2EHPA	0.3 <u>M</u>	
	TBP	0.2 <u>M</u>	
lAW (In-Line Sample	Pm-147	0.5-2 Ci/gal	Chem. Sep. followed by beta counting
	Sr-90	l Ci/gal	Chem. Sep. followed be beta counting
	Fission Products		Gamma energy analysis
	рН	4	Glass electrode - meter

TABLE XIV-3 (Cont.)

		Takimah ai	
Sample	Analysis	Estimated Concentration	Method
1AW Tk-30-2	Pm-147	0.5-2 Ci/gal	Chem. Sep. followed by beta counting
	Sr-90	l Ci/gal	Chem. Sep. followed by beta counting
	Fission Products		Gamma energy analysis
	pH .	4	Glass electrode - meter
lBP (in-line	рН	2.2	Glass electrode - meter
sample)	Sr-90		Chem. Sep. followed by beta counting
	Pm-147	0.1-0.2 Ci/gal	Chem. Sep. followed by beta counting
	Ce-144		Gamma energy analysis
	Na.	0.002-0.01 <u>M</u>	Flame photometry
	Ca	0.001-0.01 <u>M</u>	Atomic absorption
	Mg	0.001 <u>M</u>	Atomic absorption
Cone. 1BP Tk-5-1	HNO ₃	0.12-0.5 <u>M</u>	Titration with complexing
1R-)-1	Sr-90		Chem. Sep. followed by beta counting
	Na	0.1-0.2 <u>M</u>	Flame photometry
	Ca	0.01-0.1 <u>M</u>	Atomic absorption
	Mg	0.01 <u>M</u>	Atomic absorption
	Isotopic Sr		Atomic absorption
	Ce-144		Gamma Energy analy- sis
	Sr-80/Sr-90		
	Fe	0.0001-0.01 <u>M</u>	Colorimetric
	Al	0.0001-0.01 <u>M</u>	Colorimetric
	Pb	0.01 <u>w</u>	Colorimetric or atomic absorption

TABLE XIV-3 (Cont.)

Sample	Analysis	Estimated Concentration	Method
<pre>lCP-Pm (in-line sample)</pre>	Pm-1 ¹ 47		Chem. sep. followed by beta counting
	нио3	2 <u>M</u>	Titration with com- plexing
	Ce-144		Gamma energy analysis
	Persulfate	0.2 <u>M</u>	
1CW Tk-26-3	Ce-144	~-	Gamma energy analysis
Ce Prod. Tk-27-3	hno ³	<u>2M</u>	Titration with com- plexing
	Pm-147	~-	Chem. Sep. followed by beta counting
	Ce-144		Gamma energy analysis
	Al	Trace	Colorimetric
	Fe	Trace	Colorimetric
	Ag	Trace	Colorimetric or atomic absorption
	Na.	Trace	Flame photometry
	Isotopic Ce	~~ <i>**</i>	
Pm Ex-W Tk-26-2W	Pm-147	~	Chem. Sep. followed by beta counting
	Fission Products		Gamma energy analysis
	рН	3.5	Glass electrode - meter
Pm Prod. Tk-26-2P	Pm-147	400 Ci/gal	Chem. Sep. followed by beta counting
	нио	1 <u>M</u>	Titration with com- plexing
	Ce-144, Y-91		Gamma energy analysis
	sr-90		Chem. Sep. followed by beta counting
10W Tk-26-1	Sr-90		Chem. Sep. followed by beta counting
	Pm-147		Chem. Sep. followed by beta counting
	Ce-144, ZrNb-95 Y-91		Gamma Energy analysis
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TABLE XIV-3 (Cont.)

Sample	Analysis	Estimated Concentration	Method
CONC. Pm Prod. Tk-36-4	HNO ³	0.5 <u>m</u>	Titration with com- plexing
	Pm-147	10 ³ -10 ⁴ Ci/gal	Chem. Sep. followed by beta counting
	Am-241	l x 10 ¹¹ c/m/gal	Chem. Sep. followed by alpha counting
	Y-91		Chem. Sep. followed by gamma energy analysis
	Ce-144		Gamma energy analysis
	Pm-148/Pm-147		•
	Total Rare earths	0.5 <u>M</u>	
	Al	Trace	Colorimetric
	Fe	Trace	Colorimetric
	Ca.	Trace	Atomic absorption
	Ag		Atomic or colori- metric
	TMI		Emission spectro- graph

TABLE XIV-4

STRONTIUM PURIFICATION

		Estimated	
Sample	Analysis	Concentration	Method
Sr-laf Tk-29-2-Sr	рН	4.9	Glass electrode - meter
	Sr-90	PAW-600 Ci/gal PAS-100 Ci/gal	Chem. Sep. followed by beta counting
	Ne.	PAW 1.8 <u>M</u> PAS 3.2 <u>M</u>	Flame photometry
	Ca	0.01-0.04 <u>M</u>	Atomic absorption
	Ce-144		Gamma energy analysis
	Ba	Trace	Atomic absorption
	Mg	Trace	Atomic absorption
	Fe	Trace	Colorimetric
	Al	Trace	Colorimetric
Sr-lAW (In-line sample)	Sr-90	2-5 Ci/gal	Chem. Sep. followed by beta counting
	Fission products		Gamma energy analysis
	рН	5	Glass electrode - meter
Sr-1AW Tk-30-2-Sr	Sr-90	2-5 Ci/gal	Chem. Sep. followed by beta counting
	Fission products		Gamma energy analysis
	рН	5	Glass electrode - meter
Sr-1BP (In-line	рН	2.0	Glass electrode - meter
Sample)	Sr-90	PAW 700 Ci/gal PAS 500 Ci/gal	Chem. Sep. followed by beta counting
	Ce-144		Gamma energy analysis
	Citric acid	1 <u>M</u>	
Sr-PROD. Tk-5-1-Sr	Citric acid	0.05 <u>M</u>	
	нио	1 <u>M</u>	
	S r -90	10 ³ Ci/gal	Chem. Sep. followed by beta counting
	Fission products		Gamma energy
Sr-10W Tk-27-3 or 27-1	Sr-90	l Ci/gal	Chem. Sep. followed by beta counting
	Y-91		Gamma energy analysis

TABLE XIV-5

ANALYTICAL REQUIREMENTS FOR ION EXCHANGE

Sample	Analysis	Estimated Concentration	Method
PSN	Na	4 <u>M</u>	Flame photometry
105-C RSN 101-BX	Al	0.8 <u>m</u> (RSN)	Colorimetric
	Pb	Trace	Colorimetric or atomic absorption
	рH	PSN 10	Glass electrode - meter
	Cs-134, Cs-137		Gamma energy analysis
	NO3	PSN 1 <u>M</u> RSN 5 <u>M</u>	Colorimetric
	NO2_	PSN 2 <u>M</u> RSN 0.4 <u>M</u>	Colorimetric
	co ₃ -2	PSN 1M	
	so _{l4} -2	PSN 0.1 <u>M</u> RSN 0.03 <u>M</u>	
	Isotopic Cs		
1XF Tk-17-2	Na	4 <u>M</u>	Flame photometry
	рН	PSN 10 RSN 12	Glass electrode - meter
	Cs-134, Cs-137		Gamma energy analysis
1XW Tk-18-1	Cs-137		Gamma energy analysis
1XW Tk-19-1	Cs-137		Gamma energy analysis
	nh _h +	8 <u>M</u>	
	co ₃ -2	3 <u>M</u>	
	Na ·	0.01 <u>M</u>	Flame photometry
Cs Prod. Tk-20-1 Tk-14-2 Tk-17-1	Cs-137		Gamma energy analysis
	Na	0.3 <u>M</u>	Flame photometry
	Rb	0.01 <u>M</u>	Chem. Sep. Followed by atomic absorption
	Isotopic Cs		
	TMI		Emission spectro- graph
	Total Cs	0.05	Atomic absorption

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TABLE XIV-6

AQUEOUS AND ORGANIC MAKE-UP

Sample	Analysis	Estimated Concentration	Method
Solvent Tk-M0154	D2EHPA	0.3 <u>M</u>	Gas chromatography
	TBP	0.2 <u>M</u>	Gas chromatography
	SpGr.	0.8	Hydrometer
Bisulfate Tk-101	$NaHSO_{14}$	<u>6</u> <u>м</u>	
Carbonate	Na_2CO_3	2 <u>M</u>	
ISS Butt Tk-H301	HC ₂ H ₃ O ₃ (Hydroxy- acetic acid)	2.5 <u>M</u>	Qualitative test
	PН	2.4	Glass electrode-meter
Sr-1SS Butt	Citric acid	2.08 <u>M</u>	Qualitative test
Tk-H301	pН	3.4	Glass electrode-meter
Tartaric Acid Tk-H302	^н 2 ^с ^н ^н ^о 6	2 <u>₩</u>	Qualitative test
1BX-Butt Tk-H312	HNO ³	0.3 <u>M</u>	
Sr-1BX Butt Tk-H-312	Citric Acid	3.14 <u>M</u>	
Lead Carrier H-401	ьр(NO ³) ⁵	1 <u>M</u>	Indirect dichromate titration
Sugar sol'n H-403	Sugar	1.4 <u>M</u>	Specific gravity
1CX-1 27-1A	HNO3	2 <u>M</u>	
27-1B	Persulfate	0.5 <u>M</u>	
1CX-2 27-1C	HNO ₃	2 <u>M</u>	
27-1D	Ag ⁺	0.04 <u>M</u>	Thiocyanate titration
1CX 27-1A 27-1B 271-C 27-1D	(During processi	ng of sludge and 2 <u>M</u>	during Sr purification) NaOH titration
Ce Strip ^(a) 27-3-A	нио	2 <u>M</u>	NaOH titration
Ce Strip ^(b) 27-3-B	NaNO ₂	3 <u>M</u>	$\mathrm{KMnO}_{\mathfrak{t}_{\!4}}$ titration
Sr 105-2 27-3-A	hno ₃	2 <u>M</u>	

TABLE XIV-7
ANALYTICAL REQUIREMENTS FOR B-FLANT WASTE HANDLING

		Estimated	**
Sample	Analysis	Concentration	Method
HIW (Hi-level	Caustic ratio		Titration
Waste) Tk's 25-1,	S r- 90	l Ci/gal	Chem. Sep. followed by beta counting
25-2	Pm-147	2 Ci/gal	Chem. Sep. followed by beta counting
	Fission products		Gamma energy analysis
	Pu, Np, Am	Low	Chem. Sep. followed by alpha counting
LIW Tk-24-1	нио	0.1 <u>M</u>	
	Cs-137	0.0001 Ci/gal	Chem. Sep. followed by gamma energy analysis
	Sr-90	0.0001 Ci/gal	Chem. Sep. followed by beta counting
LIW Tk-23-1	рH	10-11	Glass electrode - meter
	Sr-90	0.01 Ci/gal	Chem. Sep. followed by beta counting
	Pm-147	0.02 Ci/gal	Chem. Sep. followed by beta counting
	Fission products		Gamma energy analysis
	Pu, Np, Am	Low	Chem. Sep. followed by alpha counting
	Cs-137	0.01 Ci/gal	Gamma energy analysis
	NH ¹ +	Trace	Qualitative test
RET. Basin	Gross beta	0.001 Ci/gal	Beta counting
	Fission products		Gamma energy analysis

TABLE XIV-8

ANALYTICAL REQUIREMENTS FOR STORED FISSION PRODUCTS

Sample	Analysis	Estimated Concentration	Method
CRUDE Sr	нио	0.1 <u>M</u>	Titration with com-
Tk's 6-1, 6-2 7-1, 7-2, 8-3 8-2		10 ³ Ci/gal	plexing Chem. Sep. followed by beta counting
	Na	0.2 <u>M</u>	Flame photometry
	Ca	0.01 <u>M</u>	Atomic absorption
	Mg	0.001 <u>w</u>	Atomic absorption
	Isotopic Sr		
	Fission products		Gamma energy analysis
	Sr-89/Sr-90		
	Fe	Trace	Colorimetric
	Al	Trace	Colorimetric
	Ba	Trace	Atomic absorption
Sr Prod Tk-36-1	нио	1 <u>M</u>	Titration with com- plexing
	Sr-90	10 ³ Ci/gal	Chem. Sep. followed by beta counting
	Sr Total	0.15 <u>M</u>	
	Ca	0.005 <u>M</u>	Atomic absorption
	Ва	0.003 <u>M</u>	Atomic absorption
	Mg	0.002 <u>M</u>	Atomic absorption
	Pb	0.004 <u>M</u>	Atomic absorption
	Fe	0.002 <u>M</u>	Colorimetric
	Al	0.002 <u>M</u>	Colorimetric
	Zr	0.002 <u>M</u>	Colorimetric
	Sr-89/Sr-90	0.05	
	Ce-144, Ru-106, ZrNb-95, Y-91		Gamma energy analysis
	Isotopic Sr		
	Na.	0.2 <u>M</u>	Flame photometry
	Citric Acid	Trace	

TABLE XIV-8 (Cont.)

Sample	Analysis	Estimated Concentration	Method
Pr PC	ниоз	0.5 <u>M</u>	Titration with com- plexing
Tk's 37-1, 37-2, 37-3	Pm-147	10 ¹ Ci/gal	Chem. Sep. followed by beta counting
	Am-241	10 ¹¹ c/m/gal	Chem. Sep. followed by beta counting
	Y-91	-~	Chem. Sep. followed by gamma energy analysis
	Ce-144	<u></u>	Gamma energy analysis
	Pm-148/Pm-147	10-4-10-7	
	TRE	0.5-1.0M	
	TMI		Emission spectro- graph
Pm PS (From Sludge)	HNO 3	0.5 <u>M</u>	Titration with com- plexing
- 1	Pm-147	10 ³ Ci/gal	Chem. Sep. followed by beta counting
	Am-241	10 ¹¹ c/m/gal	Chem. Sep. followed by alpha counting
	Fission products		Gamma energy analysis
	TRE	1 <u>M</u>	
	TMI		Emission spectro- graph
	Pm-148/Pm-147	10 ⁻⁷	

1421 TABLE XIV-9

ANALYTICAL REQUIREMENTS FOR 244-AR VAULT

		Estimated	
Sample	Analysis +	Concentration	Method
PAS Tk-004	Н,	0.1 <u>M</u>	Titration with com- plexing
	Sr-90	20 Ci/gal	Chem. Sep. followed by beta counting
	Ce-144, Cs-137		Gamma energy analysis
	Pm-147	10 Ci/gal	Chem. Sep. followed by beta counting
	Am, Np, Pu	Low	Chem. Sep. followed by alpha counting
	U	0.02 <u>M</u>	Fluorimetric deter- mination after chemi- cal separation
	Na	0.4 <u>m</u>	Flame photometer
	Al	0.6 <u>M</u>	Colorimetric
	Fe	0.4 <u>M</u>	Colorimetric
	Ca	0.03 <u>M</u>	Atomic absorption
Is	otopic Sr		
Sludge Slurry	рH	10	Glass electrode - meter
Tk-002	% Solids	10-20	Visual estimate after centrifugation
	% Insolubles	1	
	Cs-137		Gamma energy analysis
PAW Tk-001	H ₊	0.5 <u>M</u>	Titration with com- plexing
	% Solids	1	Visual estimate after centrifugation
	Sr-90	45 Ci/gal	Chem. Sep. followed by beta counting
	Pm-147	180 Ci/gal	Chem. Sep. followed by beta counting
	Ce-144, Cs-137		Gamma energy analysis
NHW Tk-003	Sr-90	l Ci/gal	Chem. Sep. followed by beta counting
	Pm-147	2 Ci/gal	Chem. Sep. followed by beta counting
	Cs-137, Ce-144, ZrNb-95, Ru-106		Gamma energy analysis
	Hq	10-12	Glass electrode - meter

TABLE XIV-10

ANALYTICAL REQUIREMENTS FOR IN-TANK SOLIDIFICATION FACILITY

Sample	Analysis	Estimated Concentration	Method
ITS-F 103-BY	SpGr.	1.3	Hydrometer
	% Solids	5-10	Visual estimate after centrifugation
	Cs-137		Gamma energy analysis
	Viscosity	2	
	Sr-90	0.5 µCi/gal	Chem. Sep. followed by beta counting
	Na	8 <u>м</u>	Flame photometry
	он	1 <u>M</u>	Colorimetric
	NH ₁₄ +	1 <u>M</u>	
	Reducing normality	0 to trace	
Condensate	Cs-137	 1	Gamma energy analysis
	Sr-90	10 ^{-l4} μCi/gal	Chem. Sep. followed by beta counting
	Н	9	Glass electrode - meter
	Na	0.2 <u>M</u>	Flame photometry
	Pu	10 ⁵ c/m/gal	Chem. Sep. followed by alpha counting
	Organic		Qualitative check
Salt Cake	Cs-137	500 μCi/gal	Gamma energy analysis
101-BY 102-BY	5r-90	l μCi/gal	Chem. Sep. followed by beta counting
	Melting Point		
	% Solids		Visual estimate after centrifugation
	Na	15-23 <u>M</u>	Flame photometry
	SpG	1.7	

PART VII

SAFETY

CHAPTER XV

GENERAL SAFETY

AND

HAZARDS OTHER THAN RADIATION

 $\mathbf{B}\mathbf{y}$

W. W. Roos

Personnel Protection Operation

PART VII: SAFETY (Cont.)

CHAPTER XVI

RADIATION INTENSITIES AND SHIELDING

 $\mathbf{B}\mathbf{y}$

H. H. Van Tuyl
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PART VII: SAFETY, Cont.

CHAPTER XVI: RADIATION INTENSITIES AND SHIELDING

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PART VII: SAFETY (Cont.)

CHAPTER IVI

RADIATION INTENSITIES AND SHIELDING

by H. H. Van Tuyl, BNW

A. INTRODUCTION

Large amounts of fission products and other radioactive materials must be handled in Waste Management without undue danger to the operator either from excessive exposure to the ionizing radiations or from taking the materials internally. This chapter deals with protection from external radiation, while Chapter XV deals with internal deposition of radioactive materials.

Fission products emit beta and gamma radiations. Beta radiation consists of high energy electrons. Light shielding is adequate for protection from beta radiation since it travels only a short distance through matter. Gamma radiation consists of high energy electromagnetic radiation. Massive shielding is often required for gamma radiation, especially if its energy is very high.

B. UNITS AND CONVERSION FACTORS

1. Basic Constants and Conversion Factors

Name	Symbol	
Avogadro's number	••	= 6.02252 x 10 ²³ atoms/mole (carbon- 12 scale)
Barn	= b	$= 10^{-24} \text{ cm}^2$
Curie	= Ci	$= 3.7 \times 10^{10} \text{ dis/s}$
		$= 2.22 \times 10^{12} \text{ dis/m}$
Electron Charge	= e	= 1.60210 x 10 ⁻¹⁹ Coulomb
		= 4.80298×10^{-10} $cm^{3/2}g^{1/2}s^{-1}$ (e.s.u.)
Electron Radium (Classical	= r _e	$= 2.81777 \times 10^{-13} \text{ cm}$

Electron Rest Energy = mc² = 0.511006 MeV

Electron Rest Mass = $m = 9.1091 \times 10^{-28} g$

Energy Absorbed Per Rad = 100 erg/g

Energy Absorbed Per Roentgen = 87.7 erg/g in air

= 96.5 erg/g in water or soft

tissue

Energy of One Atomic Mass Unit = 931.478 MeV

Energy of One MeV = 1.60210×10^{-6} erg

Fine Structure Constant = $= 7.29720 \times 10^{-3}$

= 1/137.0388

Natural Logarithm of 2 = $\ln 2 = 0.693147$

Planck's constant = $h = 6.6256 \times 10^{-27}$ erg sec.

Seconds in One Day = 8.64×10^{4}

Seconds in One Year = 3.1557×10^7

Velocity of Light = $c = 2.997925 \times 10^{10} \text{ cm/s}$

Watt = $w = 10^7$ erg/s

2. Activity Units

2.1 Curies

Quantities of radioactive material are most commonly expressed in curies. Originally the curie was the disintegration rate of radon-222 in equilibrium with one gram of radium, but it has since been standardized as 3.700 x 10¹⁰ disintegrations per second (or 2.22 x 10¹² disintegrations per minute). As defined the unit refers to total decay rate of a specific nuclide, regardless of decay mechanism. By extension, it is applied to the partial decay rate by a particular process. Then the unit is qualified as, for example, in alpha curie or beta curie. These are not necessarily the same as total curies. For example, americium-242 decays by beta emission and electron capture. In 100 curies of americium-242 there are approximately 810 beta curies and 190 electron capture curies.

A further extension is made when the term "gamma curie" is used as a measure of the quantity of a radionuclide. Few, if any, nuclides decay with the emission of exactly one gamma photon per disintegration. Some, such as cobalt-60, emit more than one photon per disintegration. Other nuclides, such as tellurium-125m, emit photons in being transformed to the stable or ground state of the same element. Thus, gamma curies do not necessarily relate in a simple manner to beta curies or the total number of decays. Two usages of the term "gamma curie" are common at Hanford. The first is based on the total number of gammas, while the second is based on the number of gammas of a particular energy. For dose rate purposes, only the photons which escape the atom (i.e., those which are not internally converted) should be considered. Beta curies and gamma curies should not be added together to obtain total curies. Gamma curies should be reserved for use when considering gamma detection or gamma dose rates and shielding.

An example of some differences in curie notation can be given for cerium-144 in equilibrium with its short-lived praseodymium-144 daughter. A low energy photon occurs in 13 percent of the cerium-144 disintegrations, and a 2.2 MeV photon occurs in 0.8 percent of the praseodymium-144 disintegrations, and a 2.2 MeV photon occures in 0.8 percent of the Pr-144 disintegrations. Thus, 1000 curies of Ce-144, 2000 curies of Ce-Pr-144, 1000 beta curies of Ce-144, 2000 beta curies of Ce-Pr-144, 130 gamma curies of Ce-144, and 8 curies of 2.2 MeV gamma from Ce-Pr-144 are all expressions defining about the same amount of radiation. Any of these units is acceptable provided it is adequately defined by the user.

2.2 Watts

The curie refers to the rate of disintegration without regard to energy, while the watt refers to the rate of energy released without regard to the number of atoms or rate of disintegrations. Therefore, the watt is the logical unit to use if heating is being considered. Simple application of basic conversion factors yields the equation

watts = curies x energy in MeV x 5.93 x
$$10^{-3}$$
 (1)

Consistency must be maintained between the curie and energy values. For example, if total curies are used, then the average energy per disintegration should be used. If gamma curies are used, then the average energy per photon should be used. If curies of a certain energy gamma are used, then the energy of that gamma should be used. For beta particles, the average energy should be used instead of the maximum beta energy. Conversion from maximum to average beta energy will be discussed later.

2.3 Weight

Quantities of fission products are sometimes expressed in grams of total element or in tons of uranium equivalent. These terms are not sufficiently definitive to be used by themselves in calculating dose rates. The data and methods of Chapter II can often be used to convert these indefinite quantities into values for the activity of a specific nuclide. If the weight of a nuclide is given and its half life is known, the activity can be calculated:

Ci =
$$\frac{1.3058 \times 10^8 \text{ x weight (grams)}}{\text{half life (days) x atomic weight}}$$
 (2)

Ci =
$$\frac{3.5752 \times 10^5 \times \text{weight (grams)}}{\text{half life (years)} \times \text{atomic weight}}$$

3. Dose Rate Units

3.1 Roentgen

The original dose rate unit, the roentgen, is the amount of X or gamma radiation which, in 0.001293 g of air, will produce in air ions carrying one esu of quantity of electricity of either sign. This corresponds to an energy absorption per gram of about 87.7 ergs in air or 96.5 ergs in water. Both of these values have been revised in recent years from lower values that were previously used. The value in water is higher than that in air simply because water has a higher energy absorption coefficient. The quantity of ionizing radiation incident on a detector is often calculated as a flux, ϕ , usually expressed as photons or particles per cm²·sec. Dose rate units, however, are related to the effect of the radiation on its surroundings, rather than numbers of photons or particles. A photon flux can be converted to a dose rate by simple application of conversion factors to yield the equation

$$D = 6.576 \times 10^{-5} \phi E \mu_{\rm F}/\rho \tag{3}$$

where D is in R/hr, E is in MeV, and $\mu_{\rm E}/\rho$ is the energy absorption coefficient for air in cm²/g. The energy absorption coefficient for air varies substantially with energy, as shown in Figure XVI-1. An empirical equation, good to about one percent over the range 0.01 to 10 MeV, is as follows:

$$\mu_{E}/\rho = \frac{5.0518 \times 10^{-6} E^{-3} - 4.0145 \times 10^{-3} + 0.52224 E + 0.074214 E^{2}}{1 + 12.7525 E + 7.67493 E^{2}}$$
(4)

The flux is not necessarily calculated as such, but may be implicitly included in an equation to yield dose rates. For example,

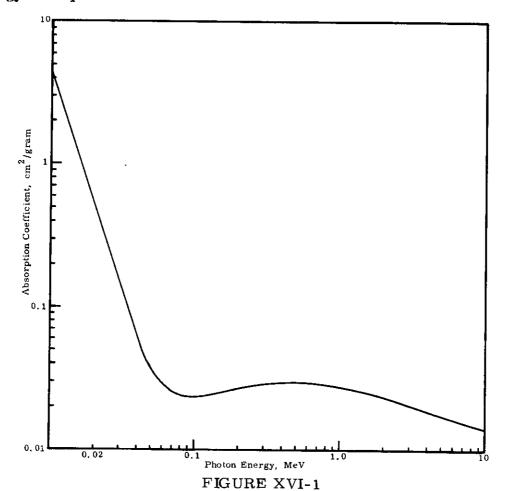
the flux at a distance r from a point source is simply the source strength in photons per second divided by the surface area of a sphere of radium r. Because of this relationship, the dose rate from a point source is inversely proportional to the square of the distance from the source. At one foot from a point source containing C curies of material, the dose rate is given by the equation

$$D = 208 C E \mu_{E}/\rho \tag{5}$$

Thus far all expressions for dose rate have involved explicit use of the energy absorption coefficient for air. Between 0.06 and 2 MeV this quantity is constant within about 20 percent. Using a value of $0.0288 \text{ cm}^2/\text{g}$ (or $3.72 \times 10^{-5} \text{ cm}^{-1}$) results in the following simplified point source conversion factor:

$$D = 6 C E \tag{6}$$

This simplified equation is adequate for most dose rate calculations, but more accuracy may be obtained by explicit inclusion of the energy absorption coefficient for air.



Energy Absorption Coefficient for Air

3.2 Rep and Rad

The Roentgen applies only to X or gamma radiation, but a similar concept is used to define dose rates from beta radiation. The Rep (Roentgen equivalent physical) is the quantity of ionizing radiation which will cause the absorption in tissue of the same amount of energy that is absorbed by one Roentgen, or about 96.5 ergs per gram. A newer unit, the rad represents 100 ergs absorbed per gram of material. The rad and Rep are often used interchangeably (in spite of the 3.5 percent error), while the Roentgen is reserved specifically for X and gamma radiation.

3.3 Rem

The Rep and rad are proper dose rate units for neutrons as well as for beta or gamma radiation. However, the biological tolerance for neutrons on an energy absorption basis is less than that for beta and gamma radiation. Thus, the Rem (Roentgen equivalent man) has been defined as the quantity of ionizing radiation which causes damage in man equivalent to one Roentgen. For fast neutrons the dose rate in Rem is about ten times the dose rate in Rep.

C. GAMMA AND X RADIATION

1. Nature and Sources

1.1 Gamma Ray

A gamma ray is a high energy electromagnetic photon resulting from nuclear rearrangements. The energy range of greatest interest in waste management is from 0.01 to 3 MeV. Gamma rays occur when an excited nuclear state de-excites to a lower energy state of the same nuclide. The simplest case, typified by 2.6 min. barium-137m, is the de-excitation of a metastable nuclear state to the ground state. In other cases, such as 58-day tellurium-125m, the metastable state may de-excite to an unstable state (i.e., less than one second half life), which then de-excites to a lower state. Thus, two or more photons may be emitted in de-exciting a single excited state of an atom. Although metastable excited states show gamma decay clearly, they are not the most important sources of gamma radiation. Most fission product gammas arise when beta decay populates an unstable excited state of the daughter nuclide. This unstable state will then de-excite by emission of one or more gamma rays until the nucleus reaches the ground state. Gamma rays also occur when an excited nucleus is formed in other ways, such as electron capture, alpha decay, fission, and other nuclear reactions.

1.2 <u>X-Rays</u>

Characteristic X-rays are different from gamma rays only in their mode of formation and range of energies. While gamma rays originate in the nucleus, X-rays originate in the extranuclear electron shells. When a K shell electron is ionized, an electron from a higher shell or from outside the atom will fill the vacancy. This filling is accompanied by the emission of quantized electromagnetic radiation (called K X-rays when the K shell is being filled) equal in energy to the difference between the initial and final energy states of the electron. The K X-ray group consists of several photon energies since electrons in several energy levels are available for filling the K shell. For convenience, the Kal X-ray energy is often taken to represent all of the K X-ray group. X-rays have energies below 0.15 MeV, while most gamma rays of interest have energies of over 0.5 MeV.

1.3 Bremsstrahlung

Both gamma rays and characteristic X-rays are emitted as monoenergetic radiation, or as a mixture of several monoenergetic radiations. Bremsstrahlung, or continuous X radiation, is emitted with energies varying from zero up to a finite high energy cutoff. This radiation occurs when a charged particle is accelerated or decelerated. The high energy cutoff is the maximum energy available for accelerating the charged particle, or the maximum energy of the charged particle being decelerated. In beta decay the radiation due to acceleration when an electron leaves the nucleus is called inner or internal bremsstrahlung, and the radiation due to deceleration of the electron is called external bremsstrahlung. In electron capture, inner bremsstrahlung is emitted, but there is no external particle to cause external bremsstrahlung. Most of the bremsstrahlung energy is concentrated in the lower energy radiations. From a shielding standpoint, bremsstrahlung is the same as gamma radiation, except that the spectrum is divided into energy groups and the total number of photons within each group is treated as a single gamma with an energy equal to the average energy of the group. Since bremsstrahlung production is intimately related to beta radiation, the calculation of bremsstrahlung spectra is described in the section on beta radiation.

1.4 <u>Internal Conversion</u>

During emission of a gamma or X-ray, internal photoelectric absorption may occur, in which the photon energy is absorbed by an extra-nuclear electron of the same atom. The kinetic energy of the emitted electron is equal to the initial photon energy less the binding energy of the electron. This binding energy is approxi-

mately equal to the energy of the X-ray for the shell from which the electron was ejected. The secondary X-ray, emitted when an electron drops into the site vacated in the conversion process, may also be internally converted. Thus, the radiation observed from gamma or X-ray transitions in a given nuclide may consist partly of monoenergetic electrons and X-rays, and fewer primary photons than expected.

The fraction of X-rays which escape internal conversion is called the fluorescent yield, ω . Values of the K shell fluorescent yield (Table XVI-1) are given approximately by

$$\omega_{K} = 1 - \frac{\mu}{E_{K}} \tag{7}$$

for values of Z above 40. The L shell fluorescent yield is lower than the K shell fluorescent yield on an atomic number basis, but about the same on an X-ray energy basis.

TABLE XVI-1

INTERNAL CONVERSION OF CHARACTERISTIC X-RAYS

Atomic Number	X-Ray Energy, KeV	Fluorescent Yield, ω
10	0.9	0.01
15	2.0	0.07
20	3.7	0.17
25	5.9	0.30
30	8.6	0.49
35	12	0.63
40	16	0.74
45	20	0.81
50	25	0.85
55	31	0.87
60	37	0.89
65	45	0.907
70	52	0.919
75	61	0.931
80	71	0.944
85	81	0.956
90	94	0.976

The extent of internal conversion of gammas is dependent on both the energy and the type of transition (i.e., spin and parity change). Conversion is very common below 0.2 MeV, and usually negligible above 1 MeV. A notable exception is the $0+\to 0+$ transition, typified by yttrium-90, which can de-excite only by internal conversion. According to most representations of the decay scheme, the de-excitation of the 1.752 MeV excited state formed by yttrium-90 decay looks like a gamma photon, but in reality no photon is emitted since only conversion electrons are allowed. The only photons from strontium-yttrium-90 decay are X-rays, bremsstrahlung, and annihilation radiation.

The extent of internal conversion has been determined experimentally for many nuclides. The result is often given as α which is the ratio of gamma conversion electrons to escaping primary photons for a specific gamma transition. Another common expression is α_K , which is the ratio of K shell conversion electrons to escaping primary photons. In this latter case, the contribution of L and higher shells should also be included in determining the number of gamma photons which leave the nucleus. Care must be exercised to see whether gamma data have already been corrected for internal conversion before applying the appropriate corrections. Many tabulations of data contain part of the gamma data corrected for internal conversion, and part of it uncorrected, with no clear indication of which is which.

A common example of the importance of internal conversion is the decay of cesium-137, which results in gamma transitions in 95 percent of the disintegrations. Ten percent of these transitions are by internal conversion instead of photon emission, so that only 85 percent of the decays give rise to gamma photons outside of the atom.

2. Attentuation Processes

2.1 Attenuation Coefficient

The most important quantity characterizing the attenuation of gamma radiation is the attenuation coefficient, μ . This quantity, which depends on the photon energy E and on the atomic number Z of the medium, may be defined as the probability per unit path length that a photon will undergo an interaction. In a thin layer, dt, within a slab shield there will occur a reduction of the intensity I of the gamma ray beam due to absorption or scattering. The resulting fractional reduction of the beam intensity, -dI/I, is proportional to the attenuation coefficient, μ , and to the layer thickness, dt; i.e.,

Integrating this equation with an incident beam intensity $\mathbf{I}_{_{\text{O}}},$ and constant $_{\mu}$ throughout the slab (homogeneous medium) gives

$$I(t) = I_o e^{-\mu t}. (9)$$

This is the basic equation for gamma or X-ray attenuation, but modifications must be made for geometry considerations and secondary radiations. The attenuation coefficient has a dimension of inverse length, and thus varies as the absorber density, This density dependence is commonly removed by use of the "mass attenuation coefficient," μ/ρ , instead of the linear attenuation coefficient. If μ is in cm $^{-1}$ and ρ is in g/cm 3 , the mass attenuation coefficient will be expressed in the customary units of cm $^2/g$. Values of mass attenuation coefficients for several elements and mixtures are listed in Table XVI-2. More complete tabulations are given by Hubbell and Berger.

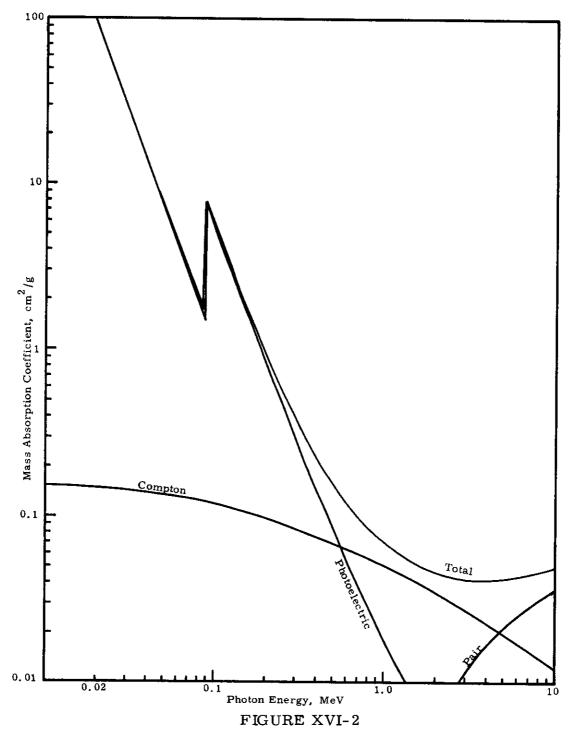
TABLE XVI-2

GAMMA RAY MASS ATTENUATION COEFFICIENTS

Mass Attenuation Coefficient, cm ² /g							
Energy,	Oxygen	Iron	Tin	Lead	Uranium	Water	Concrete
MeV	z = 8	z = 26	z = 50	z = 82	Z = 92		
0.01	5.54	179.	145.	137.	239.	4.96	23.2
0.015	1.63	57.7	46.1	114.	85.7	1.49	7.06
0.02	0.747	25.0	20.7	90.	88.8	0.705	3.01
0.03	0.332	7.91	41.9	30.6	51.5	0.335	1.00
0.04	0.230	3.46	19.5	14.3	25.2	0.243	0.495
0.05	0.195	1.80	10.5	7.96	14.5	0.211	0.326
0.06	0.179	1.11	6.48	4.72	9.11	0.196	0.254
0.08	0.162	0.554	2.95	2.12	4.42	0.178	0.193
0.1	0.151	0.347	1.63	5 .5 6	2.51	0.167	0.167
0.15	0.134	0.185	0.595	1.93	2.56	0.149	0.139
0.2	0.123	0.139	0.314	0.950	1.21	0.136	0.125
0.3	0.107	0.106	0.157	0.381	0.485	0.118	0.106
0.4	0.0953	0.0921	0.111	0.220	0.273	0.106	0.0961
0.5	0.0870	0.0830	0.0907	0.153	0.185	0.0966	0.0877
0.6	0.0806	0.0763	0.0790	0.119	0.141	0.0896	0.0811
0.8	0.0708	0.0665	0.0653	0.0853	0.0969	0.0786	0.0712
1	0.0636	0.0596	0.0572	0.0689	0.0760	0.0706	0.0640
1.5	0.0518	0.0486	0.0459	0.0510	0.0543	0.0575	0.0521
2	0.0445	0.0425	0.0407	0.0451	0.0475	0.0493	0.0449
3	0.0359	0.0361	0.0367	0.0417	0.0436	0.0396	0.0366
3 4	0.0311	0.0332	0.0357	0.0416	0.0435	0.0340	0.0319
5	0.0279	0.0316	0.0357	0.0424	0.0442	0.0302	0.0288
5 6	0.0255	0.0307	o.o 360	0.0435	0.0452	0.0276	0.0268
8	0.0226	0.0299	0.0371	0.0460	0.0479	0.0242	0.0242
10	0.0208	0.0298	0.0387	0.0491	0.0512	0.0220	0.0227

* Concrete: 1% H, 0.1% C, 52.9% O, 1.6% Na, 0.2% Mg, 3.4% Al, 33.7% Si, 1.3% K, 4.4% Ca, 1.4% Fe by weight.

For shielding purposes, the most important components of the attenuation coefficient are photoelectric absorption, Compton scattering, and electron-positron pair production. Typical values of these componenets are shown in Figure XVI-2, along with the total attenuation coefficient.



Components of the Mass Absorption Coefficient for Lead

2.2 Photoelectric Absorption

In photoelectric absorption, also called the atomic photoeffect, a photon disappears and an electron is ejected from an atom. The electron carries away all the energy of the absorbed photon, minus the energy with which the electron was bound to the atom. The Kshell electrons, which are the most tightly bound, are the most important for this effect. However, if the photon energy drops below the binding energy of a given shell, an electron from that shell cannot be ejected. Hence, a plot of the absorption coefficient versus photon energy exhibits characteristic "absorption edges." Absorption coefficient for common shield materials are shown in Figures XVI-3 and XVI-4. As a crude approximation, the photoelectric attenuation coefficient varies as the 3.5 power of the atomic number, and inversely as the cube of the photon energy. The contribution due to L and higher shells increases with atomic number from zero for hydrogen to about 25 percent of the total photoelectric attenuation coefficient for uranium.

2.3 Compton Scattering

In Compton scattering, a photon collides with an electron, loses some of its energy, and is deflected from its original direction of travel. The relationship between scattering angle and energy loss is given by

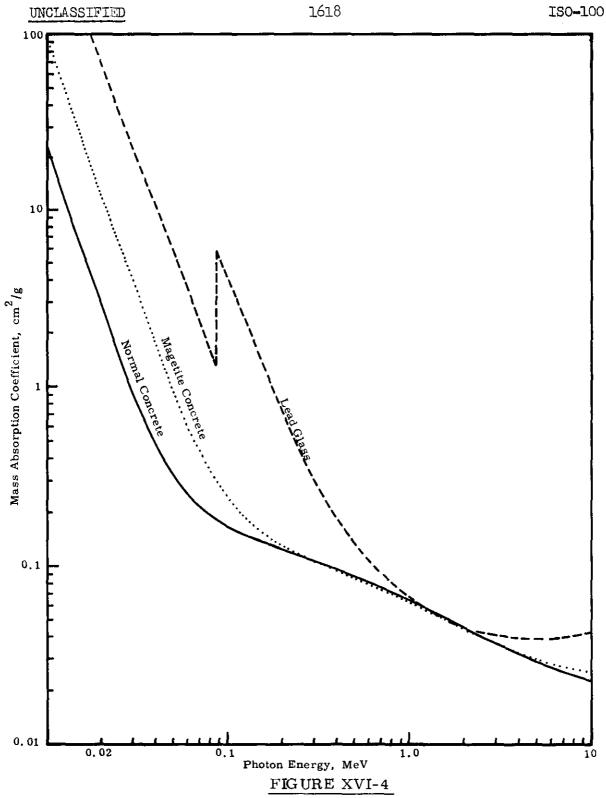
$$E' = \frac{E}{1 + (E/mc^2) (1 - \cos \theta)}$$
 (10)

where E and E' are the energies of the photon before and after the scattering, and θ is the photon deflection angle. Klein and Nishina developed an equation for the scattering probability as a function of scattering angle. Integration of this equation over all angles of scattering gives the total scattering probability, which is the Compton attenuation coefficient. Minor corrections to the Klein-Nishina theory are necessary because of electron binding effects, emission of an additional scattered photon, and certain radiation corrections. However, the Klein-Nishina equation closely approximates the actual Compton scattering probability.

Compton scattering varies with the charge to mass ration, \mathbb{Z}/A , and is thus roughly independent of atomic number. Hydrogen is a notable exception because of its unusually high charge to mass ratio.

2.4 Pair Production

In electron-positron pair production, a photon disappears in the field of a charged particle, and an electron-positron pair appears. Pair production requires a photon of at least 1.022 MeV (2 mc²) for production in the field of the nucleus, and 2.044 MeV (4 mc²) for



Mass Absorption Coefficients for Common Shield Materials (Lead Glass, Magnetite Concrete, Normal Concrete)

production in the field of an electron. Pair production occurs chiefly in the field of the nucleus, especially for high atomic number materials. The attenuation coefficient due to pair production varies approximately as the square of the atomic number.

2.5 Other Processes

In Rayleigh scattering, photons are scattered by bound atomic electrons so that the atom is neither ionized nor excited. This effect is usually disregarded in shielding calculations since little energy is lost, and scattering is either at a small angle or a small fraction of the total attenuation probability.

At higher energies (usually above 5 MeV) a photonuclear effect occurs which is similar to the photoelectric effect, but a nucleon (usually a neutron) is emitted instead of an electron. The photonuclear threshold is sufficiently high to make this process of little or no concern in Waste Management.

3. Buildup of Secondary Radiation

Each attenuation process results in some secondary photons. The photoelectric effect produces X-rays, Compton scattering produces the scattered photons, and pair production results in the 0.511 MeV radiations due to the eventual annihilation of the positron. These secondary radiations result in a higher dose rate than predicted according to the simple exponential law. This increase is usually included by use of a "buildup factor," which is the ratio of the actual dose rate to the dose rate expected in accordance with the simple exponential law. (This definition actually applies only to a dose rate buildup factor, which is the one most commonly used in shielding calculations.) The buildup factor is dependent on the gamma energy, shield material, and shield and source geometry.

The calculation of buildup factors, even for simple situations, is a formidable task since the source and shield geometries, probable energy distribution, and direction of gamma rays must be considered, and the gammas may be scattered several times before finally being removed by photoelectric capture. Factors for a few simple idealized cases, and a few different elements have been calculated by different workers, but the data currently accepted as best were calculated by Nuclear Development associates using a National Bureau of Standards, SEAC computer. Although the project required several man-years of effort, and a large amount of machine time, the results of interest to most shielding problems are quire meager in number. Some of these data, dose buildup factors for a point isotropic source in an infinite shielding medium, are presented in Table XVI-3.

TABLE XVI-3

DOSE BUILDUP FACTORS, POINT ISOTROPIC SOURCE

			Water		
μt	0.5 MeV	1.0 MeV	2.0 MeV	3.0 MeV	4.0 MeV
1 2 4 7 10 15 20	2.52 5.14 14.3 38.8 77.6 178 334	2.13 3.71 7.68 16.2 27.1 50.4 82.2	1.83 2.77 4.88 8.46 12.4 19.5 27.7	1.69 2.42 3.91 6.23 8.63 12.8 17.0	1.58 2.17 3.34 5.13 6.94 9.97
			Aluminum		
1 2 4 7 10 15 20	2.37 4.24 9.47 21.5 38.9 80.8 141	2.02 3.31 6.57 13.1 21.2 37.9 58.5	1.75 2.61 4.62 8,05 11.9 18.7 26.3	1.64 2.32 3.78 6.14 8.65 13.0 17.7	1.53 2.08 3.22 5.01 6.88 10.1 13.4
			Iron		
1 2 4 7 10 15 20	1.98 3.09 5.98 11.7 19.2 35.4 55.6	1.87 2.89 5.39 10.2 16.2 28.3 42.7	1.76 2.43 4.13 7.25 10.9 17.6 25.1	1.55 2.15 3.51 5.85 8.51 13.5 19.1	1.45 1.94 3.03 4.91 7.11 11.2 16.0
			Tin		
1 2 4 7 10 15 20	1.56 2.08 3.09 4.57 6.04 8.64	1.64 2.30 3.74 6.17 8.85 13.7 18.8	1.57 2.17 3.53 5.87 8.53 13.6 19.3	1.46 1.96 3.13 5.28 7.91 13.3 20.1	1.38 1.81 2.82 4.82 7.41 13.2 21.2

TABLE XVI-3 (Continued)

Tungsten

μt	0.5 MeV	1.0 MeV	2.0 MeV	3.0 MeV	4.0 MeV
1 2 4 7 10 15 20	1.28 1.50 1.84 2.24 2.61 3.12	1.44 1.83 2.57 3.62 4.64 6.25 (7.35)	1.42 1.85 2.72 4.09 5.27 8.07 (10.6)	1.36 1.74 2.59 4.00 5.92 9.66 14.1	1.29 1.62 2.41 4.03 6.27 12.0 20.9
			Lead		
1 2 4 7 10 15 20	1.24 1.42 1.69 2.00 2.27 2.65 (2.73)	1.37 1.69 2.26 3.02 3.74 4.81 5.86	1.39 1.76 2.51 3.66 4.84 6.87 9.00	1.34 1.68 2.43 3.75 5.30 8.44 12.3	1.27 1.56 2.25 3.61 5.44 9.80 16.3
			Uranium		
1 2 4 7 10 15 20	1.17 1.30 1.48 1.67 1.85 2.08	1.31 1.56 1.98 2.50 2.97 3.67	1.33 1.64 2.23 3.09 3.95 5.36 (6.48)	1.29 1.58 2.21 3.27 4.51 6.97 9.88	1.24 1.50 2.09 3.21 4.66 8.01 12.7

The first problem with these data is application to other shielding materials. This can be accomplished adequately by using graphs of B versus Z at the various energies and μt values for which data are available. Extension of the data to other μt and energy values is possible in various ways. One convenient way is to plot B versus μt at different energies. From this the dose buildup factor can be interpolated for any energy and μt value for the absorber chosen.

Another method of obtaining the buildup factors at different values of μt and E was developed by Taylor, who represented buildup factors as a sum of two exponentials. A modification of his method gives the equation

$$B = Ae^{\alpha \mu t} - (A-1) e^{(\alpha-0.10)\mu t}$$
 (11)

Values of the constants in this equation are chosen to fit the data of Goldstein and Wilkins. After these constants have been determined

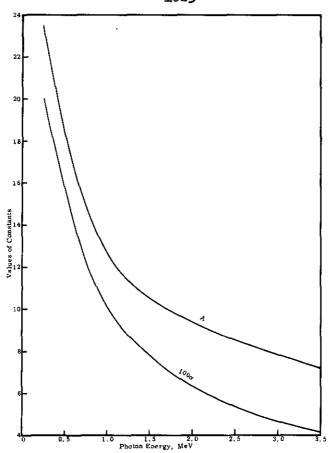
for a given absorber, energy, and range of μt values, the calculation of buildup factors is simplified for these conditions since no graphical interpolation is required. When B is expressed in this form it can be applied to an exponential attenuation equation by simply modifying the exponent. Values of these constants for water, concrete, iron and lead are shown in Figures XVI-5 to XVI-8 and in Table XVI-4.

TABLE XVI-4

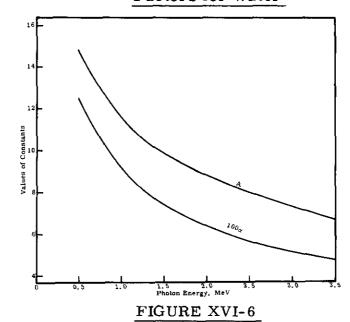
CONSTANTS FOR EXPONENTIAL REPRESENTATION

OF BUILDUP FACTORS

Energy, MeV	<u> </u>	A	Max. Error, %				
<u>Water</u>							
0.255 0.5 1.0 2.0 3.0 4.0	0.205 0.160 0.105 0.063 0.046 0.040	23.5 18.7 12.7 9.35 7.84 6.65	29 25 11.4 4.9 2.3 1.7				
	Ir	on					
0.5 1.0 2.0 3.0 4.0	0.092 0.0835 0.067 0.063 0.064	10.7 9.7 7.8 6.33 5.15	7.5 7.4 5.8 3.7 3.4				
	Lead						
0.5 1.0 2.0 3.0 4.0	0 0.0175 0.0425 0.0675 0.095	3.1 4.5 4.35 3.6 2.7	3.3 3.3 1.4 2.1 1.5				
Normal Concrete							
0.5 1.0 2.0 3.0 4.0	0.125 0.092 0.064 0.052 0.046	14.8 11.6 8.8 7.32 6.16	12.9 9.4 6.3 3.3 2.2				

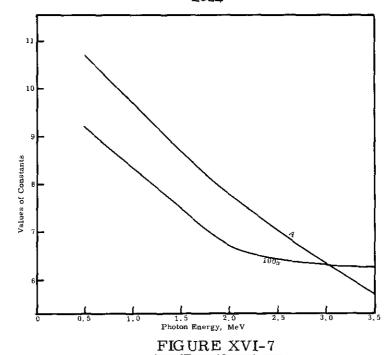


Constants for Exponential Representation of Build-Up
Factors for Water

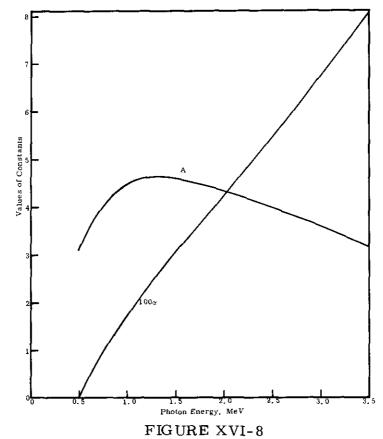


Constants for Exponential Representation of Build-Up

Factors for Normal Concrete



Constants for Exponential Representation of Build-Up Factors for Iron



Constants for Exponential Representation of Build-Up

Factors for Lead

The range of values covered by Nuclear Development Associates is adequate for most normal shielding problems, but several situations occur in which data outside this range are required. For example, shielding of bremsstrahlung from Pm-147 and shielding of gammas and X-rays from transuranic elements requires buildup factors for lower energy radiations. The calculations have, therefore, been extended by a modified "straight ahead" approximation that is much simpler (and less accurate) than the "moments" method used by Nuclear Development Associates. The two methods agreed within 30 percent over the range covered by the moments method. Results for iron and lead are shown in Figures XVI-9 and XVI-10.

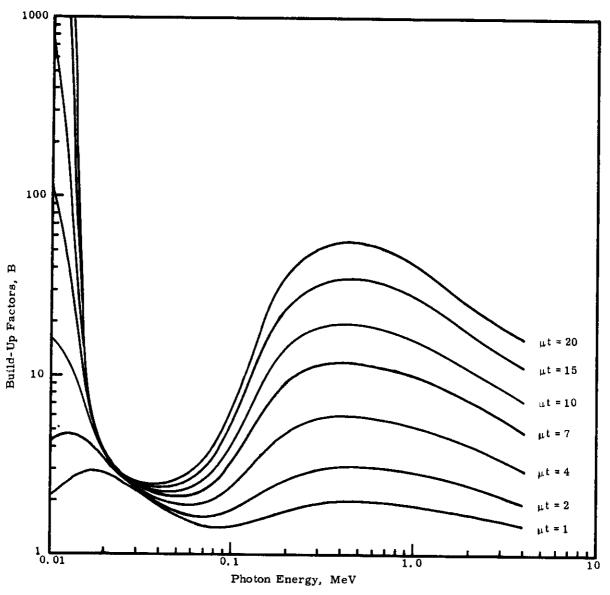
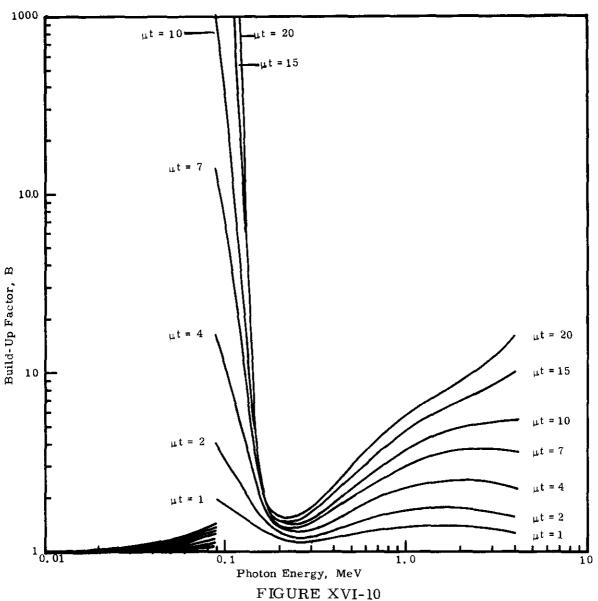


FIGURE XVI-9
Build-Up Factors for Iron



Build-Up Factors for Lead

After a maximum near 0.5 MeV the iron values decrease with decreasing energy because of the increasing importance of photo-electric absorption. A similar effect is seen with lead, but the maximum occurs at a higher energy. Another effect is seen with lead at energies slightly above the 88 keV X-ray absorption edge. The X-ray buildup factor becomes important, approaching the exponential of the difference in mean free paths for the primary photon and the X-ray. Below the X-ray absorption edge the buildup factor is again low, and decreases with decreasing energy. Selected low energy buildup factor data are shown in Table XVI-5.

162**7** TABLE XVI-5

COMPTON BUILDUP FACTORS AT LOW ENERGIES

μt	0.01 MeV	0.02 MeV	0.05 MeV	0.1 MeV	0.2 MeV		
Aluminum							
1 2 4 7 10 15 20	1.007 1.014 1.027 1.043 1.058 1.078	1.058 1.113 1.212 1.34 1.45 1.59	2.06 3.19 4.87 9.39 14.3 24.0 34.8	2.82 5.64 12.9 40.6 91.0 245.	2.71 5.35 13.2 45.3 117. 345. 718.		
			Iron				
1 2 4 7 10 15 20	1.001 1.002 1.004 1.006 1.008 1.011 1.013	1.007 1.013 1.024 1.037 1.047 1.060	1.086 1.16 1.27 1.39 1.48 1.60	1.35 1.67 2.28 3.07 3.83 5.04 6.25	1.71 2.63 4.88 9.25 14.8 24.2 37.3		
			<u>Tin</u>				
1 2 4 7 10 15 20	1.002 1.003 1.006 1.009 1.012 1.016 1.020	1.009 1.017 1.031 1.048 1.062 1.080	1.013 1.023 1.038 1.054 1.065 1.078 1.089	1.054 1.093 1.15 1.20 1.25 1.30 1.35	1.20 1.35 1.58 1.87 2.11 2.46 2.76		
		Tur	ngsten				
1 2 4 7 10 15 20	1.001 1.001 1.002 1.004 1.005 1.006	1.003 1.006 1.012 1.018 1.023 1.030	1.030 1.054 1.090 1.13 1.16 1.19	1.019 1.033 1.052 1.073 1.090 1.12 1.15	1.073 1.121 1.19 1.26 1.32 1.39		
Lead							
1 2 4 7 10 15 20	1.000 1.001 1.002 1.003 1.004 1.005	1.002 1.005 1.009 1.013 1.017 1.022	1.022 1.039 1.066 1.093 1.11 1.14	1.018 1.033 1.062 1.10 1.14 1.22	1.055 1.091 1.14 1.20 1.23 1.29		
<u>Uranium</u>							
1 2 4 7 10 15 20	1.000 1.001 1.001 1.002 1.003 1.004 1.004	1.002 1.004 1.006 1.010 1.013 1.017	1.016 1.029 1.048 1.069 1.083 1.10	1.058 1.10 1.16 1.23 1.28 1.34 1.39	1.042 1.069 1.11 1.15 1.18 1.23 1.28		

Application of buildup factors to homogeneous shields composed of different elements involves assignment of an effective atomic number to the shield. This is accomplished by determining the value of μ for the mixture, and comparing the shape of the μ versus E curve with similar curves for the elements. The atomic number of the element giving the best fit is assigned as the effective atomic number of the mixture. Buildup factors for this atomic number are then used for the composite shield.

For laminated shields where the lighter shield is on the outside, the buildup factor for the lighter shield is usually used, even though this may overestimate shielding requirements. Where the heavier shield is on the outside and μt is greater than one, the buildup factor for the heavier material only is used. For other cases the buildup factors may be weighted in accordance with the μt values in each material, or the shield may be treated as described for homogeneous shields.

The buildup factors were calculated for a point source in an infinite shield, but the error to be expected in applying these to a finite shield is usually small, especially for thick shields, high atomic numbers, and a shield perpendicular to the line between the source and detector. For angles of penetration up to about 30° the thickness of the shield is usually taken as the actual thickness traversed by straight line radiation (up to about 15 percent more shielding than the slab thickness). At greater angles either the thickness of shielding or buildup factors should be adjusted to correct for the lowered effectiveness. In most cases it is adequate to assume no greater than a 30° angle of penetration, even though the actual angle exceeds this value.

4. Geometry Effects

4.1 Symbols.

The following symbols will be used in the discussion of geometry:

- B Integrated buildup factor
- C Total gamma curies of specified energy in entire source
- D Dose rate at detector, R/hr
- D Unshielded dose rate at one foot from point source (Equation 5, R/hr
- E Photon energy in MeV
- a Distance from detector to nearest point of source, feet

b ut value for shield, or sum of ut values for layered shield

b' μ r value for source (used only for three dimensional sources such as cylinders and spheres)

- h Height of cylinder, feet
- r Radius of disc, cylinder, or sphere, feet
- s Length of line source, feet
- t Perpendicular thickness of slab shield, feet
- μ Absorption coefficient for external shield, feet $^{-1}$
- μ Absorption coefficient for source, feet⁻¹

Values of exponential and secant integrals have been either tabulated or plotted in several books on radiation shielding. A few values are included in Figures XVI-11 and XVI-12. For large values of b, and n \geqslant 1, $E_n(b)$ may be evaluated by

$$\frac{e^{-b}}{b+n} < E_n(b) \leq \frac{e^{-b}}{b+n-1}$$

4.2 Point Sources

All of the previous discussion has assumed a point source of radiation. Even though this condition is never actually met, it is by far the most used and the easiest to calculate. Other simple finite geometries are the line, plane disc, cylinder, and sphere. Calculations for some of these geometries are possible by integration of formulas for point sources to yield relatively simple analytical expressions. Most other geometries yield expressions which are best evaluated by computers rather than manual methods. Solid figures, such as the cylinder and sphere, involve consideration of shielding within the source (self-shielding) as well as the effects of distance on dose rate. The most important simple geometries are discussed below, but more extensive compilations of both exact and approximate methods for other geometries have been published.

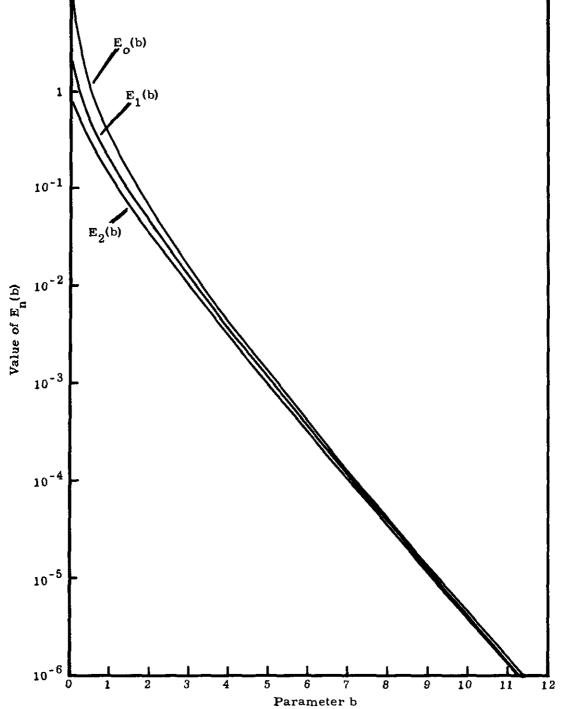


FIGURE XVI-11
Exponential Integral Functions

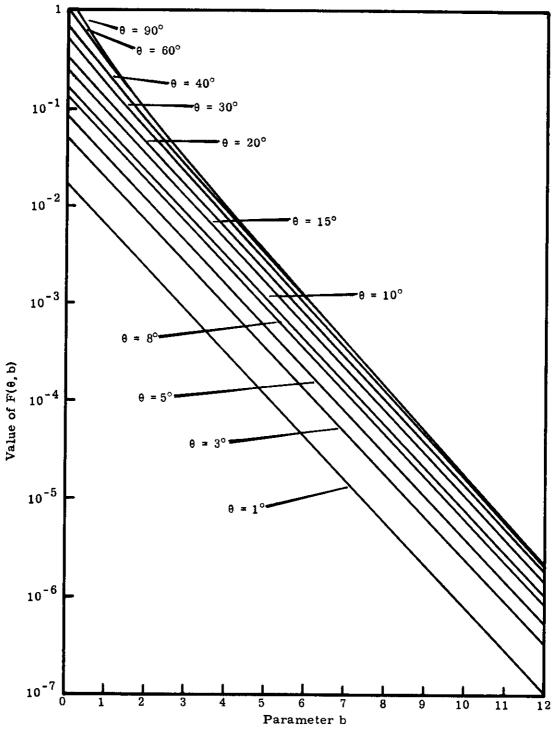


FIGURE XVI-12
Secant Integral Functions

The elementary formula for dose rate from a point source of radiation is the basis for all of the other formulas in this section:

$$D = \frac{B D_o}{a^2} e^{-b}$$
 (12)

The buildup factor, B, given by Equation 11 may be substituted to obtain the following equation:

$$D = \frac{D_0}{2} [A e^{-b(1-\alpha)} - (A-1) e^{-b(1.1-\alpha)}]$$
 (13)

This equation is particularly valuable when evaluating buildup for sources other than point sources. For a point source the buildup factor is evaluated for a constant shield thickness, but for other sources the shield thickness varies as different points of the source are considered. This variation of buildup factor can be included by using a simple point attenuation kernel for integration, and evaluating the result by first substituting $b(1-\alpha)$ for b, and then substituting $b(1-\alpha)$ for b. A combination of the two results, using the appropriate constant multiplier for each, will properly account for buildup, corrected for variation in shield thickness with location in the source.

4.3 Line Sources

The line source approximates liquids in tubing or pipes, ion exchange, or solvent extraction columns, or in general any cylinder where the radius is small compared to the distance between the source and detector. If self-shielding is important, the line source approximation should not be used.

Three cases can be profitably distinguished with the line source and slab shield: I. A line parallel to the shield; II. A line perpendicular to the shield but not passing through the detector; and III. A line perpendicular to the shield and passing through the detector. In the first case, a simplification can be made by considering only a line beginning at the perpendicular from the detector to the line of the source, and extending in one direction. Other arrangements with this can can be handled by addition or subtraction of the appropriate line sources.

Case I.
$$D = \frac{B D_o}{as} F(tan^{-1} \frac{s}{a}, b)$$
 (14)

Case II.
$$D = \frac{B D_0}{sc} [F(tan^{-1} \frac{c}{d}, b) - F(tan^{-1} \frac{c}{d+s}, b)]$$
 (15)

where c is the perpendicular distance from the detector to the line of the source, and

Case III.
$$D = \frac{B D_0 e^{-b}}{a(a+s)}$$
 (16)

It is possible to approximate the radiation from a line source by considering the slab shield thickness to be constnat. For Case I, this gives

$$D = \frac{B \ D_{o}}{as} \ (e^{-b}) \ (tan^{-1} \ \frac{s}{a}). \tag{17}$$

This approximation gives a dose rate about a factor of (1 + 0.05b) too high at an angle of 30° for b up to 20, but the error is smaller at smaller angles. For sources which end a distance c from the perpendicular from the source to the slab, b should be replaced by

 $\sqrt{\frac{ba}{a^2-c^2}}$ to compensate for the increased thickness of shielding. For closer approximations, the line source may be divided into a series of line sources, each of which subtends a rather small angle at the detector.

For Case II, the constant slab thickness approximation gives

$$D = \frac{B D_0}{sc} (e^{-b})(tan^{-1} \underline{c} - tan^{-1} \underline{c})$$
 (18)

where c and d are defined for this case in the solution given above, and b may be replaced by ab.

4.4 Disc Sources

The disc source approximates contamination on flat surfaces, or cylinders of small height where self-shielding in the source is negligible. For a disc source observed on the axis of the disc (the line perpendicular to the disc and passing through the center of the disc), and with the slab parallel to the disc, a simple analytical expression is possible.

$$D = \frac{2 B D_0}{r^2} [E_1 (b) - E_1 (\frac{b}{a} \sqrt{a^2 + r^2})]$$
 (19)

When the shielding is considered to be constant (for example, at small angles or at angles over 30°),

$$D = \frac{B D_0}{r^2} e^{-b} \log(\frac{a^2 + r^2}{a^2})$$
 (20)

This approximation yields an error of about a factor of (1 + 0.1b) for an angle of 30° .

When the detector is not on the axis of the disc, or when the shield is not parallel to the disc source, evaluation is much more difficult.

4.5 Cylindrical Sources

Cylindrical sources are probably the most common actual sources encountered in shielding calculations. Here not only the distance from volume elements of the source to the detector and the actual thickness of slab shielding, but also the shielding of the source itself should be considered. Since the limits of the triple integration are not simple expressions, exact solutions are not possible. An exact solution is possible for a truncated cone viewed on the axis of the cone, and this solution can be used to give upper and lower limits for the simple case of a cylinder with the axis perpendicular to the shield. The dose rate from a cylinder is less than that from a truncated cone with the top of the cone equal to the base of the cylinder,

$$D < \frac{2B D}{rhb} \left[\mathbb{E}_{2}(b) - \cos \theta \mathbb{E}_{2} \left(b \sec \theta \right) - \mathbb{E}_{2} \left(k \right) + \cos \theta \mathbb{E}_{2}(k \sec \theta) \right]$$
(21)

where
$$\tan \theta = \frac{r}{a}$$
 and $k = \frac{1}{r}(rb + h b') = b + \frac{h}{r}b'$

The dose rate is more than that from a truncated cone with the base equal to the base of the cylinder,

$$D > \frac{2 B D_{o}}{rhb!} [E_{2}(b) - \cos \theta E_{2}(b \sec \theta) - E_{2}(k) + \cos \theta E_{2}(k \sec \theta)]$$
where $\tan \theta = \frac{r}{a+h}$ and $k = \frac{1}{r}$ (rb + hb!).

For other orientations of a cylinder on end, or for the cylinder axis parallel to the shield, the calculations are even more complicated.

4.6 Spherical Sources

The first two integrations for a spherical source and slab shield can be readily performed, but the final integration is more difficult:

$$D = \frac{1.5 \text{ B D}_{o}}{r^{2}b'} \int_{0}^{\theta} \left[1 - \exp\left(-2b' \sqrt{1 - \csc^{2} \theta \sin^{2} \psi}\right)\right] e^{-b \sec \psi} \sin \psi d \psi(23)$$
where $\sin \theta = \frac{r}{s+r}$.

This final integration can be performed graphically or numerically. Another possibility is to replace part of the expression by a simple approximation. Since the first exponent varies from -2b' to 0, using a constant value of -2b' will give a conservative estimate of the dose rate. Using this approximation and integrating gives:

$$D < \frac{1.5 \text{ B D}_{0}}{r^{2} \text{b'}} (1-e^{-2b'})[E_{2}(b) - \cos \theta E_{2}(b \sec \theta)]$$
 (24)

Another approximation, especially useful and accurate for small values of b', involves integration of the first exponential by approximating a term of sec ψ e as e. This approximation is conservative for values of b above 1 for θ up to 30°, and for even smaller values of b at smaller angles. This integration yields:

$$D = \frac{3 B D_o e^{-b}}{8(b')^3 (a + r)^2} [2(b')^2 - 1 + (2b' + 1) e^{-2b'}]$$
 (25)

A more convenient approximation is obtained by regarding $\sec \psi$ as 1, and the entire first exponential as 0. In the integrated equation, the function $(1-\cos\theta)$ may be approximated adequately as $1/2\sin^2\theta$ for small angles. Using all of these approximations gives:

$$D = \frac{3 B D_{o} e^{-b}}{4 (a + r)^{2} b'}.$$
 (26)

This differs from the formula for a point source at the center of the sphere only by a factor of 0.75/b' and from equation 25 by the b' term. It differs from equation 25 by only 30% at b' = 1, 13% at b' = 2, and less at higher values of b'. It agrees with equation 24 at b = 20, b' above 1.5 within 25% for θ = 10° , a factor of 2 for θ = 20° , and a factor of 3 for θ = 30° . The agreement is much better at smaller values of b (e.g., 35% at b = 5, θ = 30°). Therefore, although this formula is extremely simple, it is quite accurate for a wide range of shielding conditions.

5. Approximate Methods For Calculating Dose Rates and Shield Thickness

Preliminary evaluation does not require detailed shielding calculations, but adequate accuracy is provided by techniques for obtaining quick, order-of-magnitude estimates of dose rates and shield thicknesses. These methods are based on point source geometry with buildup factors included in the calculations.

5.1 Unshielded Dose Rate

The unshielded dose rate may be obtained from Equation 12, using either Equation 5 or 6 to evaluate D depending on the accuracy desired. A nomogram for obtaining unshielded dose rates by Equations 5 and 12 is shown in Figure XVI-13. A straight edge can be laid across columns A and B at the known values of source strength (gamma curies) and gamma ray energy, respectively, and the intersection point on reference column C determined. By laying the straight edge between this reference point and column D at the known distance to the detector, the intersection point on column E determines the unshielded dose rate at the detector in mR/hr. If the source strength falls outside the range of the scale in column A, it may be brought into range by dividing by some power of 10, represented by 10 on the nomogram. The resultant dose rate obtained from the nomogram must then be multiplied by the same power of 10 to yield the correct dose rate.

5.2 Shielded Dose Rate

The attenuation of the dose rate in passing through a shield of thickness t is given by the attenuation factor, Be $^{-\mu t}$. Values of attenuation factors for point sources and slab shields with several common shielding materials are shown in Figures XVI-14 to XVI-19. This attenuation factor must then be multiplied by the dose rate at the detector calculated without any intervening shielding.

5.3 Shield Thickness

If an estimate of required shield thickness is desired, the desired dose rate after shielding is divided by the dose rate from Figure XVI-13 to determine the required attenuation factor. An estimate of shielding requirements can then be obtained from the graphs in Figures XVI-14 to XVI-19 by reading the shield thickness corresponding to the required attenuation factor and gamma energy level. Conversely, if the shield thickness is known, the figures can be used to determine the attenuation factor of the shield. By multiplying the attenuation factor by the unshielded dose rate obtained from Figure XVI-13, an estimate of shielded dose rate at a detector can be obtained.

These figures have been used to calculate dose rates as a function of shield thickness for several fission products of interest and lead shields, as shown in Figures XVI-20 and XVI-21. The dose rate obtained from these curves must be multiplied by the source strength in kilocuries and divided by the square of the distance in feet between source and detector.

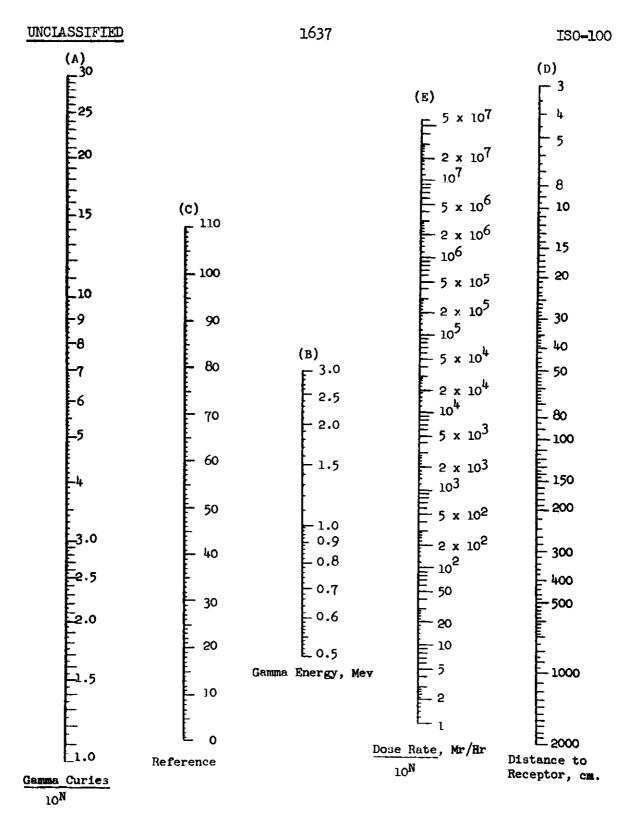
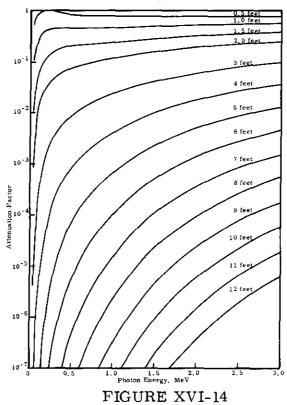
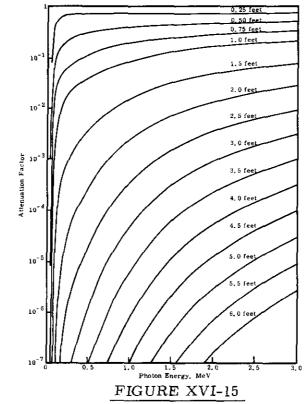


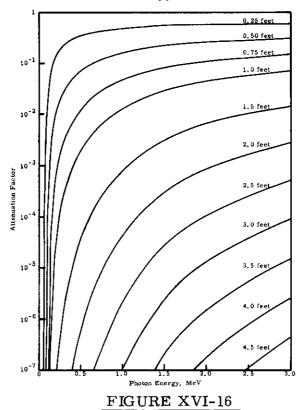
FIGURE XVI-13
Unshielded Dose Rate From Point Source



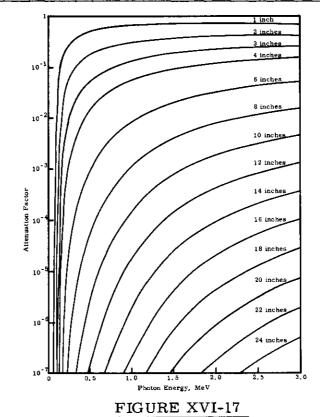
Attenuation Factors for Water, Density 1.0



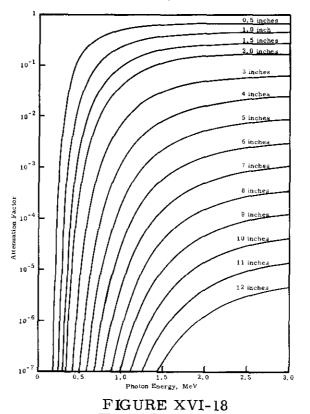
Attenuation Factors for Normal Concrete, Density 2.3



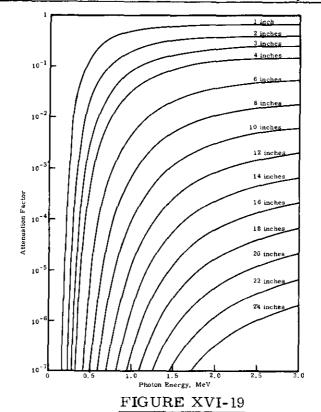
Attenuation Factors for Magnetite Concrete, Density 3.5



Attenuation Factors for Iron, Density 7.85



Attenuation Factors for Lead, Density 11.2



Attenuation Factors for Lead Glass, Density 6.2

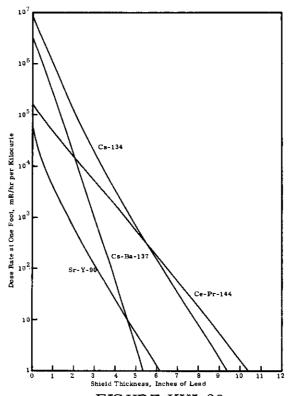
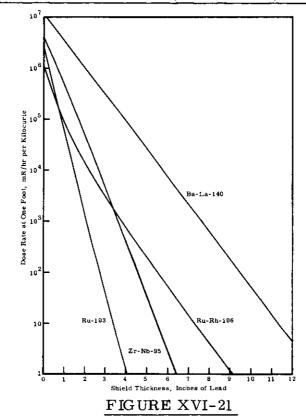


FIGURE XVI-20
Dose Rate from One Kilocurie Point Sources
Shielded with Lead (Sr-Y-90, Cs-Ba-137, Ce-Pr-144)



Dose Rate from One Kilocurie Point Sources Shielded With Lead (Zr-Nb-95, Ru-103, Ru-Rh-106, Ba-La-140)

6. Scattered Radiation and Sky Shine

Gamma radiation travels in a straight line, but interaction with a scatterer causes secondary radiation to be emitted in all directions. The effect of scattered radiation along the path of the primary gamma causes buildup, as previously discussed. Radiation from Compton scattering at greater angles has lower energies and lower scattering probabilities, but is still present. Thus, radiation which impinges on a shield at a steep angle of incidence can "turn the corner" to proceed through the shield normal to its surface as scattered radiation. Therefore, as previously mentioned, shield thicknesses should be considered as no greater than 15 percent more than the perpendicular thickness of the shield.

Radiation can also scatter or reflect from walls or even from air to go around shadow shields or labyrinths designed to shield out direct line radiation. As a crude approximation, each 90° turn that the radiation makes will be accompanied by a tenfold decrease in dose rate. The increased path length for the scattered radiation will also improve attenuation by the inverse square law. An accurate treatment of scattered radiation is much too complex for inclusion in this manual, but may be obtained from several of the sources listed in the bibliography.

7. ISOSHLD Computer Code

Most shielding calculations are either laborious or approximate. Hand calculations are prohibitively time-consuming, except for the rather simple geometries described previously, and repetitive calculations for even these simple geometries is time-consuming. Computer program ISOSHLD was developed to fill the need for a convenient semirigorous calculational method to handle the majority of these problems.

The program was designed to include most of the basic data required for these calculations. The data, contained in libraries chained to the code, include attenuation coefficients, gamma ray decay energies and probabilities, maximum beta energies and probabilities, fission product data, and buildup factor data. For most problems the user need only supply 1) the geometry and material composition of the source and 2) the geometry and materials of the shield. Optional modes of data entry are available for solving special problems.

The solution of radioisotope shielding problems often requires consideration of the amount and distribution of bremsstrahlung produced by the slowing down and stopping of beta particles. Indeed, problems are often encountered in which a mixed emission of beta and gamma rays must be treated. Therefore, a revised version

of the code (ISOSHLD-II) has the added capability of solving shielding problems for which bremsstrahlung may contribute all or part of the penetrating radiation. Bremsstrahlung is produced only in the source region; i.e., the beta particles are completely stopped by the material in that source region.

The point kernel integration technique is used to perform the attenuation calculation so that the dose at the exposure point is the contribution from a large number of individual point sources. A numerical integration is carried out over the source volume to obtain the total dose.

The code obtains solutions for any combination of the variables listed below:

Shield and Source Geometry

- 1. Point
- 2. Line
- 3. Sphere
- 4. Sphere with slab shields
- 5. Truncated cone
- 6. Disc
- 7. Cylinder
- 8. Cylinder with slab shields
- 9. Cylinder end
- 10. Rectangular solid
- 11. Infinite slab and plane
- 12. Exponential source distribution where applicable

Isotope Selection from Calculated Fission Products

- 1. Noble gases
- 2. Halogens
- 3. Volatile solids
- 4. All except the above 3
- 5. All fission products
- 6. Individual isotopes by individual specification

Source Type

- 1. Calculated source strength from known fuel irradiation exposure (see Note).
- 2. Specify curies of isotopes in library--both fission products and activation products.
- 3. Source strength in photons of specific energies for source volume.

Note: If Source Type 1 is specified, the irradiation history of the fuel must be described by supplying:

- 1. Initial conversion ratio
- 2. Fuel exposure time
- 3. Fast fission in U-238 per thermal fission
- 4. Reactor power history
- 5. Cooling time
- 6. Proper choice of above variables also approximates fuel irradiation in a fast reactor.

Shield Region Geometry and Materials

- 1. Number of regions up to five
- 2. Material in each region choice of 20 materials in library
- 3. Material density in each region five materials allowed in a region
- 4. Region and material for which buildup is most important.

D. BETA RADIATION

1. Nature and Sources

Most of the primary fission products are unstable with respect to beta decay since they contain an excess of neutrons. A high energy electron called a beta particle is emitted from the nucleus, thus converting a neutron to a proton, and bringing the nuclide closer to stability. Simultaneously with the beta particle, a neutrino is emitted. The sum of the energies of the beta particle and neutrino is equal to the total decay energy available for that particular beta transition. The amount of energy given to either particle may vary from zero to the maximum available energy, thus yielding beta particles with a continuous spectrum of energy values. The neutrino can be neglected since it is extremely penetrating and escapes without interacting with the surroundings. It is of no practical concern except for its role in causing the continuum of beta particle energies. The beta particle, however, is important because of biological, radiolytic, and heating effects.

A few nuclides in spent reactor fuels can decay by converting a proton to a neutron. This can be accomplished by positron emission, which is very similar to beta decay except for the charge on the electron. After leaving the nucleus, the positron is annihilated by an electron to yield two photons of 0.511 MeV gamma radiation. The maximum kinetic energy of the positron is 1.022 MeV less than the decay energy available. If the maximum decay energy available is less than 1.022 MeV, positron emission is impossible, but a proton can capture an extranuclear electron and thus be converted to a neutron. Usually a K shell electron is captured, giving this process the name of electron capture of K capture. It is unlikely that either positron emission or electron capture will be of any real concern in Waste Management.

1.1 Beta Spectra

Two types of beta transitions based on spin and parity changes are of greatest interest in the study of fission product beta spectra. The "allowed" transitions involve a spin change of 0 or 1, and no parity change (0 or 1, no); while the "unique first forbidden" transitions involve a spin change of 2 and a change of parity (2, yes). Spectra for transitions of these types can be calculated, but spectra for most other transitions cannot be simply and accurately predicted as yet.

The shape of the energy spectrum of beta particles is dependent on several factors, especially the decay energy, atomic number of the nucleus, and transition type. Typical beta spectra calculated for Z=50 are shown in Figure XVI-22. The shape of the energy spectrum of beta particles is used to determine the average energy of beta particles for use in heat calculations, and to determine the energy distribution of bremsstrahlung.

The relative number of betas, P(E), of kinetic energy E from and emitter of atomic number Z and maximum kinetic energy E is given for an allowed transition by:

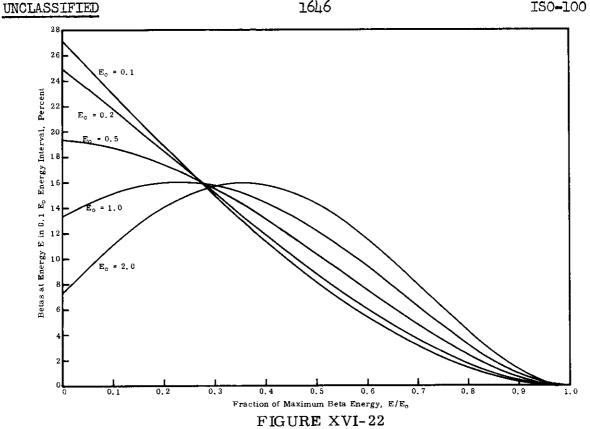
$$P(E) = K_{p}WF(Z,W)(E_{o} - E)^{2}$$
 (27)

where K is an arbitrary constant, W is the beta total energy in rest mass units, p is the beta momentum, $W^2 - 1$, and F(Z,W) is the Fermi differential function. Values of the Fermi function have been tabulated by the National Bureau of Standards as $p^2F(Z,W)$ where their Z is for the product nucleus. A screening correction such as the one calculated by Reitz should be applied to the NBS values. Since the correction usually is quite small, only an approximate evaluation of it is required.

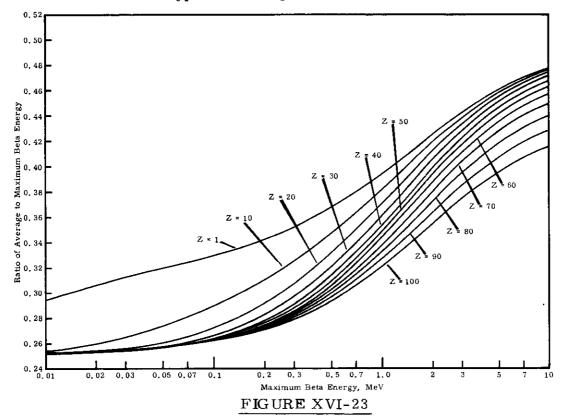
Distributions for unique first forbidden transitions are given by:

$$P(E) = K pW F(Z,W) (E_Q - E)^2 p^2 + (W_Q - W)^2$$
 (28)

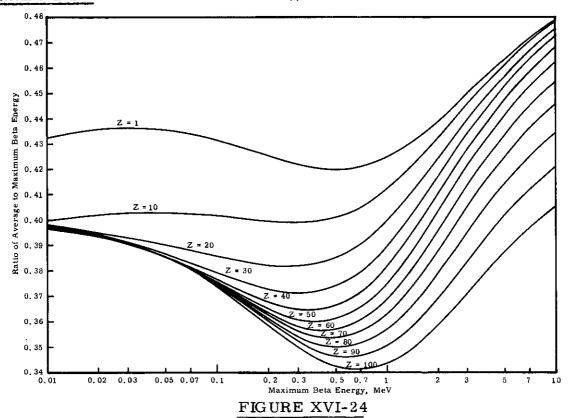
The average beta energy is approximately one-third of the maximum beta energy, but it varies from about 25 to 50 percent of the maximum energy, depending on the atomic number, energy, and transition type. For some purposes the approximate average energy is adequate, but more accurate values may be obtained easily. The ratio of average to maximum beta energy is shown in Figure XVI-23 for allowed transitions, and in Figure XVI-24 for unique first forbidden transitions. The average beta energy determined from these curves can then be used in calculating heat generation by beta decay.



Typical Beta Spectra for Z = 50



Average Beta Energy for Allowed Transitions



Average Beta Energy for Unique First Forbidden Transitions

1.2 Bremsstrahlung

An electron that is accelerated or decelerated emits part of its energy as electromagnetic radiation. Radiation emitted when a beta particle leaves a nucleus is called internal bremsstrahlung; when a beta particle is absorbed, the radiation is called external bremsstrahlung. In either case the spectral distribution is from zero to the maximum beta energy, but most of the energy is in the less energetic radiations. Even though all beta emitting nuclides produce bremsstrahlung, substantial interest in this process as applied to beta emitters has developed only recently. The nuclear gamma radiation from most radionuclides overshadows bremsstrahlung radiation. The importance of bremsstrahlung radiation in handling beta emitters was first widely recognized when highly purified strontium sources were prepared and used in thermoelectric generators, and shielding had to be provided for the bremsstrahlung radiation. Since that time the necessity of shielding for bremsstrahlung from other beta emitters has also been recognized.

The energy emitted as inner bremsstrahlung depends only on the beta energy and is small even for very energetic electrons. For the 1.5 MeV beta of Y-91 this accounts for only about 0.16 percent of the total beta energy.

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External bremsstrahlung increases with increasing beta energy and increasing atomic number (Z) of the absorber. The ratio of energy loss by radiation to that by ionization for a monoenergetic electron is approximately

$$\frac{(d E/dX)rad}{(d E/dX)ion} = ZE/800$$
 (29)

where E is the kinetic energy of the electron in MeV. Although this approximation is rather inaccurate for low energy electrons, it is usually adequate because the low penetration of bremsstrahlung from low energy electrons usually does not make great accuracy imperative. Application of this formula to beta emission requires division of the beta spectrum into several energy groups and calculation of the total bremsstrahlung energy resulting from each group. This gives only the total amount of bremsstrahlung and not the energy spectrum. For each energy group of betas, the maximum bremsstrahlung energy is equal to the maximum energy of betas in that group.

An approximate bremsstrahlung spectrum S(k), for photons of energy k may be evaluated as the product of three factors, summed over all beta energies from k to E_0 . The factors are 1) the fraction of the total beta energy at energy E, 2) the fraction of this energy which is lost by radiation, and 3) the fraction of this radiated energy which has energy k. The total energy of bremsstrahlung of energy k per Δk photon energy interval per unit beta energy is thus given by:

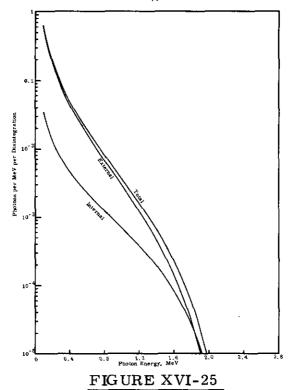
$$S(k) = \sum_{E=k}^{E} \frac{E P(E)}{\Sigma E P(E)} \frac{ZE}{ZE + 800} \frac{1}{1.25E} \left[4(1 - \frac{k}{E}) + 3\frac{k}{E} \ln \frac{k}{E} \right] \Delta k \quad (30)$$

where
$$\Sigma \to P(E)$$
 is from $E = 0$ to $E = E_0$.

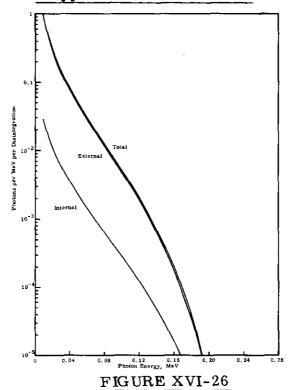
More accurate evaluations of both internal and external brems-strahlung spectra are possible with computer code BREMRAD, or with the methods and equations given in several of the references. Results of these more accurate calculations for strontium-yttrium-90 and for promethium-147 are shown in Figures XVI-25 and XVI-26, respectively.

2. Beta Dose Rates and Shielding

For most applications to beta particles the problems of dose rate and shielding need to be considered together since even the source itself provides significant attenuation of the betas. Calculations with betas are generally much less important than with gamma rays because of the much lower penetrating power of the betas, but they may become important at Hanford since pure beta emitters are being isolated.

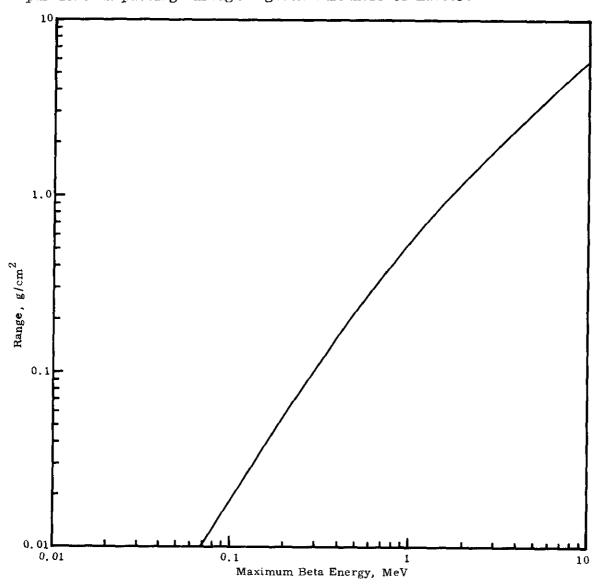


Bremsstrahlung Spectra from Sr-Y-90 Betas stopped in Strontium Oxide



Bremsstrahlung Spectra from Pm-147 Betas stopped in Promethium Oxide

Beta particles passing through matter interact with electrons in the matter to lose their energy by ionization. The energy loss per unit weight of absorber is reasonably independent of atomic number, since the number of electrons per unit mass determines the stopping power, and this varies as Z/A. Because of the large number of collisions between beta particles and the electrons of the absorber, the range of betas with a given energy and absorber is virtually constant. The range shown in Figure XVI-27 is specifically for aluminum, but is useable for any absorber by inclusion of corrections for the number of electrons per unit mass. Even without this correction the range curve is a good approximation. This range curve is useful in both dose rate and shielding calculations since it can be used to determine the energy lost by an electron or beta particle in passing through a given thickness of matter.



Range of Beta Particles in Aluminum

Beta particles above about 0.5 MeV undergo about 15 collisions per cm of air, and cause about 60 ion pairs per cm, which corresponds to an energy loss of about 2 KeV per cm. The ionization loss per cm increases as the beta energy decreases. An approximate value of the specific energy loss (energy loss per path length) may be obtained by dividing the beta energy by the total range, but this is somewhat in error because of the higher ionization at lower energies. This value, together with the geometry, can be used to determine the dose rate. For point source geometry, and neglecting shielding, the dose rate is approximately

rads/hr at one foot = $1.8 \times 10^5 \times \text{energy} \times \text{curies/range}$ (31) where the energy is in MeV and the range is in mg/cm². This formula actually applies to monoenergetic electrons rather than to the spectrum of beta energies, but is a reasonable approximation.

An example of shielding calculation is the amount of steel required to completely shield a strontium-yttrium-90 source. The maximum beta energy is 2.27 MeV, which corresponds to 1.37 g/cm² of aluminum. Because of the Z/A correction this is equal to about 1.41 g/cm² of steel. If the density of steel is taken as 7.8 g/cm³, this is 0.18 cm, or 0.071 inches. Thus, slightly more than 1/16 inch of steel will completely shield the beta radiation from strontium-yttrium-90. Shielding for bremsstrahlung must be handled the same as for gamma radiation.

An example of dose rate calculations is the dose rate one foot from one curie of strontium-90 and its yttrium daughter, with the source being weightless but enclosed in 1 mm of glass. The weight of shielding is 0.1 cm x 2.6 g/cm³ = 260 mg/cm² of glass, plus 30.5 cm x 1.29 mg/cm³ = 39.4 mg/cm² of air. This shielding will absorb all beta particles of energy below about 0.63 MeV, and will reduce the yttrium-90 maximum beta energy from 2.27 MeV to about 1.8 MeV, with a residual range of the beta being about 1070 mg/cm2. Because of the absorbers the strontium-90 beta will not contribute to the dose rate at one foot. Since the average energy of yttrium-90 betas is about 0.88 MeV, slightly less than half of these betas are absorbed. The spectrum of beta energies one foot from the source is essentially from 0 to 1.8 MeV with a distribution similar to the yttrium-90 beta distribution from 0.63 to 2.27 MeV. The average energy of this spectrum is about 0.7 MeV, with a range of 260 mg/cm². Using these values, and a source strength of 0.6 Curies to allow for absorption, the dose rate is approximately 300 rads/hr. The source self-shielding and geometry factors should usually be included in a practical problem, but are omitted here for simplicity.

E. ALPHA RADIATION

Many nuclides, particularly above mass 200, are unstable with respect to emission of a helium nucleus, or alpha particle. The energy liberated in alpha decay is usually between 5 and 7 MeV, with most of the energy going to the alpha particle. Some energy also appears as recoil energy of the emitting nuclide. The alpha emitting nuclides most likely to be encountered in Waste Management are uranium-235, uranium-238, plutonium-238, plutonium-239, plutonium-240, neptunium-237, americium-241, curium-242, and curium-244. The alpha particles have a very short range and are of little concern in shielding, except for their role in producing neutrons as discussed below. Gamma radiation or X-rays accompany the decay of most alpha emitters. These radiations are treated the same as gamma or X-rays from beta emitters.

F. NEUTRONS

Fast neutrons are emitted in the process of spontaneous fission, which occurs with several heavy nuclides such as plutonium-238, plutonium-240, curium-242, and curium-244. The neutron generation rate due to spontaneous fission depends only on the amount of the fissioning nuclide present, and is not affected by the chemical form or composition.

Another source of fast neutrons is the reaction of alpha particles with light elements by an α ,n reaction. Beryllium yields a particularly high number of neutrons in this way, but other elements also produce a significant number of neutrons. Table XVI-6 lists the expected dose rates due to α ,m neutrons with several different light elements mixed with selected alpha emitters. The dose rate from α ,n neutrons thus depends on the type and amount of light elements associated with the alpha emitters.

TABLE XVI-6
NEUTRON DOSE RATES FROM ALPHA EMITTERS

Light	Relative	Dose Rate, mrem/hr at one foot per gram					
<u>Element</u>	Effectiveness	Po ²¹⁰	Pu ²³⁸	Am ²⁴¹	Cm ²⁴²	Cm ²⁴⁴	
D.	1000	110 000	500	7.00	000 000	1000	
Be	1200	112,000	500	100	200,000	4200	
C	1.7	140	0.7	0.14	260	5.6	
0	1.0	88	0.4	0.08	160	3.4	
F	100	8,800	40	8.0	16,000	340	
Al	12	1,100	5.0	1.0	2,000	42	
Sponta	neous Fission		0.03		190	100	

Neutrons from α ,n reactions generally have a higher energy and a smaller energy spread than do neutrons from spontaneous fission. However, attenuation of neutrons of the two types is roughly comparable.

Fast neutrons are very penetrating in most materials, but they are slowed down or moderated by light elements, especially hydrogen. After moderation, they may be stopped by many materials. Figure XVI-28 shows the attenuation of neutron dose rates by water and by lithium hydride shields as a function of shield thickness.

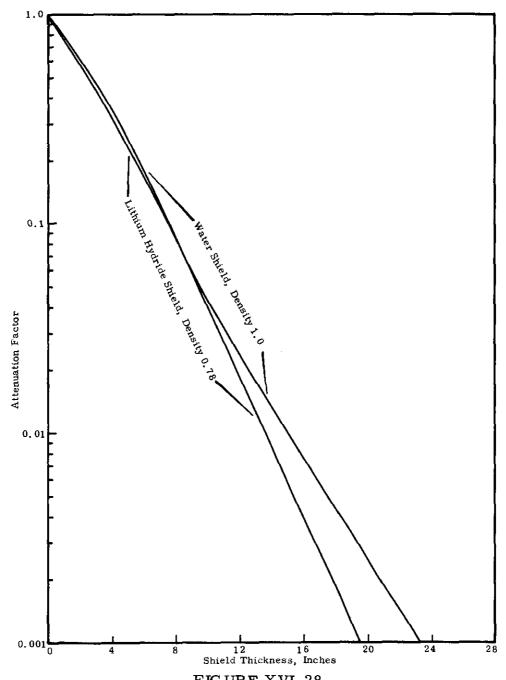


FIGURE XVI-28
Attenuation of Dose Rate From Fast Neutrons

The expected neutron dose rate at one foot from one gram of selected alpha emitter oxides is shown in Table XVI-7 for different thicknesses of water shielding. The neutron dose rates are directly proportional to the weight of alpha emitter and inversely proportional to the square of the distance from the source to the dose point, at least for distances over one foot.

TABLE XVI-7
NEUTRON ATTENUATION BY WATER SHIELDING

Shield Thickness,	Dose Rate, mrem/hr at One Foot Per Gram							
Inches	Po ²¹⁰	Pu ²³⁸	Am ²⁴¹	Cm ²⁴²	Cm ²⁴⁴			
THERES								
0	88	0.43	0.08	350	103			
ž ‡	33	0.16	0.030	133	39			
8	7.8	0.038	0.0071	31	9.1			
1 2	2.2	0.011	0.0020	8.7	2.6			
16	0.67	0.003	0.0006	2.7	0.78			
20	0.22	0.001	0.0002	0.9	0.26			

When neutrons are absorbed penetrating gammas sometimes result. These gammas may be more difficult to shield than the original neutrons. The effect of secondary gammas has been included in the above discussion, but may be altered if additional elements are present in the moderator.

Neutron dose rates will probably be of no importance in Waste Management unless alpha emitters are recovered in a concentrated form.

G. NUCLEAR DATA

Dose rate and shielding calculations require an accurate knowledge of the penetrating radiations associated with the source material. Chapter II contains information for obtaining the activities in mixed fission products in terms of total curies, but does not provide information on beta or gamma energies and abundances. Translation from total curies to individual beta and gamma energies and abundances requires a knowledge of the decay schemes involved. Table XVI-8 contains such data for principal fission products, and Table XVI-9 contains data for selected alpha emitting nuclides. The gamma data have been corrected for internal conversion, and conversion electrons (if appreciable) have been included with the beta particles. Brems-strahlung radiations are not included in the table.

Most of the data were derived from Nuclear Data Sheets and the Journal of Nuclear Data, updated where applicable by the 9th edition of the KAPL Chart of the Nuclides. Some more recent decay scheme investigations are also included.

TABLE XVI-8
SIMPLIFIED DECAY SCHEMES OF FISSION PRODUCTS

			Particles		Photons	
Nuclide	Half Life	Max. E, MeV	Avg. E, MeV	Abund., %	E, MeV	Abund., %
Se 79	6.5x10 ⁴ y	0.160#	0.060	100		
Kr 85	10.6 у	0.672 * 0.15	0.252 0.041	99.6 0.42	0.514	0.42
Rb 86	18.7 d	1.777 * 0.696	0.723 01232	91 9	1.084	9
Sr 89	50.6 d	1.463*	0.582	100	0.913	0.01
Sr 90	28 y	0.544*	0.201	100		
Y 90	64 h	2.27 * 0.52 * 1.74 ce	0.942 0.191 1.74	100 0.017 0.017	0.511 pair 0.016 Kx	0.0045 0.011
¥ 91	59 đ	1.54 * 0.33	0.615 0.097	99.7 0.3	1.208	0.3
Zr 93	9.5x10 ⁵ y	0.063 * 0.034 *	0.024 0.013	75 25		
Nb 93m	3.7 y	0.026 ce 0.010 ce	0.026 0.012	88 12	0.017 Kx	9
Zr 95 (Nb 95m)	65 d (90 h)	1.13* 0.885* 0.396 0.360	0.437 0.335 0.120 0.107	0.4 2 55 43	0.760 0.726 0.235 0.017 Kx	43 55 0.8 0.7
Nb 95	35 d	0.93 * 0.16 0.748 ce	0.352 0.044 0.748	1 99 0.2	0.768	99
Tc 99	2.1x10 ⁵ y	0.292*	0.107	100		
Ru 103 (Rh 103m)	40 d (57m)	0.71 0.39 0.212 0.10 0.475 ce 0.037 ce	0.234 0.117 0.059 0.026 0.475 0.037	3 1 89 7 0.6 91	0.610 0.498 0.362 0.297 0.040	7 89 0.3 0.4 0.25
Ru 106 (Rh 106)	1.0 y (30 s)	0.0394 3.54 3.0 2.4 2.0 1.5 1.3 1.2	0.010 1.51 1.26 0.977 0.792 0.564 0.477 0.434	100 78 8 11 2 0.5 0.2 0.4 0.09	2.9 M 2.4 M 2.0 M 1.6 M 1.1 M 0.9 M 0.624 0.612	0.012 0.026 0.09 0.27 2.4 0.7 9.9 1
Pd 107	7x10 ⁶ y	0.035*	0.014	100		
Cd 109 (Ag 109m)	453 d (40 s)	0.085 ce 0.062 ce	0.085 0.062	52 44	0.088 0.022 Kx 0.003 Lx	14 100 10

TABLE XVI-8 (Cont.)

TABLE XVI-8 (Cont.)						
			Particles		Photons	
Nuclide	Half Life	Max. E., MeV	Avg. E, MeV	Abund., %	E, MeV	Abund., %
Ag 110 m (Ag 110)	260 d (24 s)	2,87 0.53 0.085	1.19 0.166 0.022	2 33 65	1.5 M 1.384 0.937 0.885 0.76 M 0.69 M 0.656	18 24 34 72 32 35 93
Ag 111	7.5 d	1.05 0.79* 0.69	0.370 0.292 0.226	92.7 1.1 6.2	0.342 0.247	6 1
Cd 115 m	43 d	1.63* 0.687* 0.335 0.20	0.644 0.252 0.098 0.055	96.7 2 1 0.3	1.30 1.14 0.935 0.485	1 0.03 2.3 0.3
Sn 123 m	129 d	1.42 * 0.34	0.551 0.100	98 2	1.08	2
SD 124	60 a	2.313 1.59 1.016 0.954 0.621 0.225 0.051	0.932 0.601 0.355 0.330 0.199 0.063 0.013	23 5 1.5 5 51 11 2	2.09 M 1.69 1.4 M 1.0 M 0.72 M 0.645 0.603	6.3 48 10 4 14 7.2 98
Sn 125	9.6 đ	2.33* 1.3* 0.47 0.37	0.953 0.50 0.144 0.109	95 1.3 1.5 2.1	1.97 1.068 0.904 0.811 0.468	1.2 3.9 0.9 1.0 0.5
Sb 125	2.7 y	0.612* 0.437 0.295 0.233 0.124 0.118 0.09	0.223 0.133 0.085 0.065 0.033 0.031	13.4 5.8 40.4 1 30.7 6.0 1.9	0.668 0.633 0.60 M 0.43 M 0.35 M 0.205 M 0.174 M	1.6 11 24 41 2 0.5 7.1
Te 125 m	58 d	0.105 ce 0.078 ce 0.030 ce	0.105 0.078 0.030	47 53 13	0.1096 0.0353 0.027 Kx	0.3 7.5 114
Sn 126 (Sb 126 m)	2x10 ⁵ y) (19 m)	0.4	0.12 0.74	100 1	0.695 0.665 0.415	1 1 1
Sb 126	12.5 d	1.9	0.74	100	0.695 0.665 0.415	100 100 100
Te 127 m (Te 127)	105 d (9.3 h)	0.73* 0.695 0.27 0.054 0.084 ce 0.057 ce	0.268 0.226 0.077 0.014 0.084 0.057	1.5 97.5 1 0.013 56 42	0.665 0.418 0.360 0.21 M 0.089 0.059 0.027 Kx	0.013 0.83 0.12 0.1 0.1 0.5 37 CLASSIFIED

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TABLE XVI-8 (Cont.)

Particles Photons							
<u>Nuclide</u>	Half Life	Max. E., MeV	Avg. E, MeV	Abund., %	E, MeV	Abund., %	
Te 129 m (Te 129)	33 d (67 m)	1.59 1.453 0.99 0.7 0.3 0.101 ce 0.074 ce	0.601 0.542 0.343 0.23 0.09 0.101 0.074	5 76 15 3 1 47 48	1.12 0.72 0.475 0.211 0.106 0.027 0.027 Kx	1 16 2 0.3 25 41	
I 129	1.6x10 ⁷ y	0.150 * 0.035 ce 0.005 ce	0.055 0.035 0.005	100 17 73	0.040 0.030 Kx	10 64	
I 131	8.05 d	0.81* 0.606 0.33 0.25	0.298 0.193 0.096 0.070	0.7 87.5 9.1 2.8	0.724 0.638 0.364 0.284 0.2 M 0.08	2.8 9.1 80 5.2 2	
Xe 131 m	12 d	0.159 ce 0.129 ce	0.159 0.129	35 63	0.164 0.030 Kx	2 55	
Cs 134	2.1 y	1.453* 0.892* 0.658 0.410 0.282 0.086	0.561 0.329 0.211 0.122 0.080 0.022	0.2 1.5 61 9 0.5 27	1.57 1.365 1.0 M 0.800 M 0.605 0.566 M 0.475	0.1 5 8 83 98 25	
Cs 135	2 x 10 ⁶ y	0.210*	0.076	100			
Cs 136	13 d	0.657 0.341	0.211 0.099	7.4 92.6	1.255 1.065 0.830 0.337 0.2 M	21 84 100 59 67	
Cs 137 (Ba 137 m)	30 y (2.6 m)	1.18 * 0.514 * 0.625 ce	0.446 0.185 0.625	5 95 10	0.6616 0.032 Kx	85 9	
Ba 140	12.8 d	1.01 0.9* 0.6 0.48 0.025 ce	0.349 0.33 0.19 0.15 0.025	60 5 10 25 77	0.537 0.43 M 0.304 0.16 M 0.030	25 6 4 9 16	
La 140	40 h	2.20 1.71 1.38 1.10 0.83	0.873 0.653 0.506 0.387 0.276	7 10 45 26 12	2.9 2.52 1.597 0.9 M 0.8 M 0.45 M 0.329	0.1 6 94 15 28 43 20	
Ce 141	32.5 d	0.580 0.435 0.138 ce 0.103 ce	0.183 0.131 0.138 0.103	30 70 3 19	0.145 0.036 Kx	48 17	

TABLE XVI-8 (Cont.)

	TADIM NAT-O (OOMO)								
Nuclide	Half Life	Max., E., MeV	Avg. E, MeV	Abund., %	E, MeV	Abund., %			
Pr 143	13.8 d	0.933	0.318	100					
Ce 144 (Pr 144)	285 d (17 m)	2.98 2.29* 0.80 0.32 0.24 0.186	1.222 0.918 0.263 0.092 0.067 0.051	97.7 1.3 1 76 4 20	2.18 1.49 0.691 0.134 0.100 0.080	0.8 0.25 1.6 10.5 0.1			
Pm 146	5.53 у	0.78 0.145	0.256 0.039	33 2	0.74 M 0.633 0.589 0.454	58 2 0.6 65			
Nd 147	11.1 d	0.82 0.50 0.38 0.22	0.271 0.153 0.112 0.061	65 2 28 5	0.65 M 0.532 0.43 M 0.3 M 0.14 M 0.091	2.3 25 6 7 2 27			
Pm 147	2.62 y	0.225 0.104	0.0605 0.027	100 0.01	0.121	0.004			
Pm 148 m (Pm 148)	41 d (5.4 d)	2.48* 1.93 1.02 0.69 0.50 0.40	1.002 0.741 0.350 0.221 0.152 0.118	3.8 1 3.5 24 18 52	1.465 0.95 M 0.727 0.630 0.605 M 0.551 0.4 M	2.4 41 36 87 14 94 46			
Sm 151	90 y	0.0759 0.0543 0.020 ce 0.014 ce	0.020 0.014 0.020 0.014	98.4 1.6 0.5 1.0	0.0216 0.007 Lx	0.08 0.5			
Eu 152	12.4 y	1.484 1.05 0.70 0.38 0.20	0.542 0.359 0.224 0.111 0.055	6 1 12 3 2	1.409 1.25 M 1.10 M 0.96 M 0.779 0.45 M 0.3 M 0.122	25 8 28 18 13 7 31 30			
Eu 154	16 y	1.855 0.87 0.58 0.26	0.701 0.288 0.180 0.073	10 24 38 28	1.61 1.276 1.00 M 0.874 0.724 0.593 0.248 0.123	2 38 30 15 20 6 6 31			

TABLE XVI-8 (Cont.)

Particles Photons						
Nuclide	Half Life	Max., E., MeV	Avg. E, Me	Abund., %	E, MeV	Abund., %
Eu 155	1.8 y	0.247 0.185 0.158 0.140 0.097 ce 0.078 ce 0.055 ce	0.069 0.050 0.043 0.037 0.097 0.078 0.055	15 10 32 43 0.8 2 8	0.1053 0.0865 0.0600 010453 0.0265 0.0188	21 33 1.7 0.8 1 0.03
		0.036 ce 0.010 ce	0.036 0.010	13 40	0.043 Kx 0.007 Lx	32 10
Eu 156	15 d	2.447 1.21 0.48 0.30	0.069 0.419 0.145 0.085	32 11 34 23	2.18 M 2.05 M 1.90 M 1.25 M 1.0 M 0.86 M 0.812 0.65 M	5.3 11.5 5 33 13 2 9.5
ть 160	72.4 d	1.72 0.86 0.56 0.46 0.26	0.640 0.284 0.172 0.138 0.073	0.4 31 36 19 14	1.27 M 1.18 M 0.966 M 0.879 0.75 M 0.36 M 0.30 M	13 18 35 31 3 2 27
Тъ 161	6.9 d	0.584 0.509 0.452 0.215	0.182 0.155 0.135 0.059	10 64 26 0.1	0.300 0.10 M 0.074 M 0.057 M 0.049 0.026 0.046 Kx	0.1 0.4 10 5 19 21 26

Nuclide identification and half lives in parenthesis refer to short-lived daughters included in equilibrium with long-lived parent.

^{*} Beta with unique first forbidden shape. Others have allowed shape.

ce Conversion electron instead of beta particle.

M Multiple gammas near the listed photon energy.

Kx K x-rays from internal conversion or K capture.

Lx L x-rays from internal conversion or L capture.

Pair Annihilation radiation resulting from internal conversion by positron-electron pair creation.

TABLE XVI-9
PROPERTIES OF SELECTED ALPHA EMITTERS

	Po ²¹⁰	Pu ²³⁸	Am ²¹ 41	Cm ²⁴²	Cm ²⁴⁴
Half Life, years	0.379	89	458	0,446	18
Specific Activity, Ci/g	4500	17	3.24	3320	81
α Energy, MeV	5.30	5.49	5.48	6.10	5.79
Spont. Fission Half Life		4.9x10 ¹⁰	2x10 ¹⁴ y	7.2x10 ⁶	1.4x10 ⁷
Neutron Generation, n/g-sec Spont. Fission a,n as Oxíde	1.1x10 ⁷	3.4x10 ³ 5.4x10	1.0x10 ¹	2.3x10 ⁷ 2.0x10 ⁷	1.2x10 ⁷ 4.2x10 ⁵
Selected Gammas*	0.803 (1.2-3)	0.013 (28) 0.044 (0.038) 0.760 (5-5) 0.875 (2-5)	0.013 (90) 0.060 (36) 0.618 (1.1-4) 0.661 (7.0-4) 0.688 (1.0-4) 0.721 (2.8-4)	0.013 (26) 0.562 (1.8-4) 0.605 (1.4-4) 0.89 (2.5-5) 1.01 (1.0-5)	0.013 (23) 0.100 (1.5-3) 0.150 (1.3-3) 0.610 (2.5-5) 0.820 (6.5-5) 0.863 (8.3-6)

^{*}First value is energy in MeV. Value in parentheses is photons per 100 decays. Values such as (1.2-3) and (5-5) represent a number and a power of ten; i.e., 1.2x10-3 and 5x10-5 respectively. Spontaneous fission and fission product gammas are not included in the tabulation.

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PART VII, Continued SAFETY

CHAPTER XVII

HEALTH PROTECTION FROM RADIATION

Ву

W. N. Koop

Personnel Protection Operation

PART VII: SAFETY, Cont. CHAPTER XVII

HEALTH PROTECTION FROM RADIATION

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CHAPTER XVII - HEALTH PROTECTION FROM RADIATION

This chapter presents key features of radiation safety programs established to protect not only employees but people located beyond areas of Waste Management operations from the hazardous radiations resulting from those operations. These programs are summarized in the ARHCO Manual of Radiation Protection Standards and Controls. Since the standards and controls are frequently reviewed and revised, they should be considered prime reference sources.

There is no discussion in this chapter on biological effects of radiations nor upon the consequences of losing control of either radiation exposure or of contamination. Other sources of information are available on these pertinent subjects. Furthermore, facility design is not discussed here, since the subject is treated extensively in other portions of this manual. It must be acknowledged however, that facility design plays a major role in achieving effective radiation protection. Design engineers have followed approved radiological criteria in Waste Management facilities with consequent reductions in radiation exposure and improvement in contamination control. The presence of radiation protection personnel on the design council assures that radiation protection principles will be consistently applied in subsequent facility modifications.

A. NATURE OF RADIATIONS

Materials processed in Waste Management operations are radio-active. The radiations produced are from atomic nuclei as they change spontaneously from an unstable condition to a more stable form by the process of radioactive decay. Nearly all the beta and gamma radiations in these facilities are from the decay of fission products. Where alpha radiation is found, the source is the decaying atoms of uranium or of transuranic elements. Alpha and beta radiations consist of high velocity charged particles in contrast to gamma radiation that consists of electromagnetic waves of the same general type as light and radio waves but characterized by extremely short wave lengths.

1. Alpha Particles

An alpha particle, with 2 protons and 2 neutrons, has a mass of four, a positive change of two, and is structurally identical to the helium nucleus. Because of the charge and large mass, even high velocity alpha particles (5 MeV) have only a range of about 3.5 cm in air or 0.004 cm in tissue. A thin sheet of paper, a film of water, or the outer layer of skin will completely stop alpha particles. Because of this limited penetration, bodily damage from external alpha radiation is negligible. However, because alpha particles produce an extremely dense path of ionization before being stopped (one alpha particle may produce 150,000 ion pairs), it is vitally important to prevent alpha emitters from entering the body. Significant internal exposure is sustained from relatively small depositions of any alpha emitting material.

2. Beta Particles

Beta particles are electrons emitted from decaying radioactive atoms. They have energies up to 12 MeV or speeds almost to that of light. Particles of more than 2 MeV are rare, however, in waste management facilities where the prime sources of beta radiation are aged fission products. The distance a 2 MeV beta particle travels is limited to less than 700 cm in air, .9 cm in water or tissue, and .08 cm in lead. Because such a particle penetrates to the germinal layer of the skin, external exposure from beta radiation can be significant. Furthermore, beta particles can cause external exposure indirectly by bremsstrahlung or secondary photon radiation (X-rays) producted by deacceleration of the electrons passing through matter. However, the prevention of beta emitters entry into the body tends to dominate protection efforts.

3. Gamma Rays

Gamma rays are a form of electromagnetic radiation that have wave lengths of about one-millionth that of light. They are emitted from the nucleus of some atoms undergoing radioactive decay in packets or photons of discrete energies. Energy is lost exponentially as it traverses through matter and so the rays are not considered to have a definite limit or range. Gamma rays are very penetrating, much more so than either alpha or beta radiations. Since Chapter XVI deals more completely with the attenuation of gamma radiation, only a brief example will be cited here. The intensity of a 2 MeV gamma ray beam is reduced to 0.2 MeV after passing through 1.8 inches of lead or 2.7 inches of steel or 19 inches of water. The same reduction of intensity for this energy gamma ray would require 1300 feet of air. Within waste management facilities the presence of gamma radiation is the prime source of external exposure.

B. OCCUPATIONAL PROTECTION

An effective program of radiation control is based on the limits of radiation exposure suggested by recognized knowledgeable bodies. The permissible radiation exposure limit is the quantity of radiation which under any combination of circumstances is not expected to cause appreciable bodily injury to the average normal individual at any time during his lifetime.

1. Definition of Terms

1.1 Roentgen

A roentgen (R) is the quantity of X or gamma radiation such that the associated corpuscular emission per 0.001293 gram of air produces in air, ions carrying one electrostatic unit of quantity of electricity of eigher sign. (NOTE: 0.001293 gram is the mass of one cubic centimeter of dry atmospheric air at 0°C and 760 mm Hg pressure.) This unit of dose corresponds to the absorption of about 83.8 ergs/gram of air.

1.2 Rad

The term rad is an acronym for radiation absorbed dose. A rad is 100 ergs of absorbed energy per gram of absorbing material. The rad is a measure of the energy imparted to matter (i.e. retained by matter) by ionizing radiation per unit mass of irradiated material at the place of interest.

1.3 Rem

A rem (roentgen equivalent man) is that quantity of any ionizing radiation such that the energy imparted to a biological system (cell tissue, organ or organism) per gram of living matter by the ionizing materials present in the region of interest has the same biological effectiveness as an absorbed dose of one rad of X-radiation (about 0.4 MeV) in the same region. The use of the rem permits adding exposures from mixed radiations by recognizing the relative biological effectiveness of the various radiations. The list below is used for this purpose.

X-rays or gamma rays 1R → 1 rad = 1 rem beta particles 1 rad = 1 rem alpha particles 1 rad = 20 rem

1.4 Curie

A curie (Ci) is defined as that quantity of a radioactive material disintegrating at the rate of 3.7 x 10^{10} atoms per second and is the unit used in measuring radioactivity of these materials. Since the curie is a relatively large unit, the millicurie (mCi = 0.001 Ci) and the microcurie (μ Ci = 10^{-6} Ci) are often used. The curie is based on the number of nuclear disintegrations and not on the number of radiations emitted. For example, some radionuclides emit two or more gamma photons for each nuclear disintegration and hence the decay scheme or the exact mode of decay must be known if accurate values of activity are to be calculated from experimental data. Furthermore, due to counting efficiencies being much less than 100 percent, conventional measuring instruments do not count all radiations emitted, so corrections need to be applied to the counts recorded.

2. Exposure Limits in Controlled Areas

The program to control radiation exposure is designed to conform to the intent of AEC Manual Chapters 0230, 0524 and 0525. Limits of exposure are established in the ARHCO Manual of Radiation Standards and Controls and are summarized below.

Type of Exposure	Period	of Time	Dose (rem)
Whole body, head and trunk, blood forming organs, gonads, lens of the eye	Lifetime Calendar Calendar	•	5 (n-18)* 12 3
Bone, thyroid, skin	Calendar Calendar	•	30 10
Other single organ	Calendar Calendar	_	15 5
Skin of hands, forearms, feet and ankles	Calendar Calendar		75 25

*Where N is the age in years.

The above standard limits exposure sustained from radiation sources both internal and external of the body. In a rigorous evaluation of sustained exposure against these limits, external dose and internal dose are additive. In many routine protection programs in the nuclear industry, exposures from external sources up to the limit are allowed, while avoiding significant depositions of radionuclides (internal dose). This practice is followed in the Waste Management facilities where depositions of radionuclides can usually be prevented but where accurate determinations of dose from radionuclide depositions can be very difficult. In contrast, external exposure can be measured with adequate accuracy and relative ease. Routine control, therefore, involves careful measuring of external exposure and preventing bodily entry of radioactive materials through wounds, ingestion or inhalation.

The measurement of external radiation exposure is achieved primarily by the use of individually assigned beta-gamma film badge dosimeters. These dosimeters are processed rapidly and the results promptly made available to management. The film badge dosimeter measurement is supplemented, as required, by measurements made with other individually assigned dosimeters, (i.e., film finger rings and "pencils") and by measurements made with fixed and portable instruments that are discussed later in this chapter.

Bioassay programs and whole body counter examinations are used to assess radiation exposure from internally deposited radionuclides. The frequency and type of measurement are established on the basis of the exposure potential of the individual's work assignment, and is dependent upon the physical and biological properties of the materials with which he works.

Radiation exposure records for each worker are maintained in individual folders. All results obtained from the external and internal exposure

measurement program are periodically transferred to these folders, along with the evaluations and interpretations of these results. In addition to the routine information, any unusual incident in which the individual may have been involved or any unusual exposure he may have received is investigated, documented and placed in the individual's record file. References to the medical treatment or work restrictions which are occasionally required as a consequence of an incident are also retained in this file.

3. Radiation Zones and Radiation Work Procedures

A Radiation Zone is established where significant radiation exposure can be received. The following criteria are applied in establishing Radiation Zones:

- 1) Where there is a persistent radiation dose rate in excess of 1 mrem/hour, or
- 2) A potential radiation dose rate in excess of 50 mrem/hour, or
- 3) Smearable surface contamination is likely to be present in amounts exceeding 200 c/m beta-gamma contamination per 100 square centimeters or 500 d/m alpha contamination per 100 square centimeters, or
- 4) Fixed surface contamination exceeds 1000 c/m beta-gamma or 500 d/m alpha contamination from plutonium or 2000 d/m alpha from natural uranium, per 100 square centimeters, or
- 5) Where airborne contamination potentially can exceed the 40-hour occupational maximum permissible concentration.

In posting Radiation Zones, no distinction is made as to the nature of the hazard (i.e., external radiation, surface contamination or airborne contamination) which is present, nor any radiation protection controls which may be required. However, any entry to a Radiation Zone and the performance of all work in the zone is in accordance with a "Radiation Procedure" established for the location and work where whole body dose rates greater than 25 rem/hour can be encountered. Access is often controlled on a physical basis by lock and interlock systems in addition to administrative access control.

Radiation Procedures detail the requirements for protective clothing, respiratory equipment, personnel monitoring devices, surveillance of work by Radiation Monitoring, the locking out of process equipment, etc. There are many different Radiation Procedures used and while

some authorize work for only one shift most of them are prepared for use over an extended period. Radiation Procedures require approvals of operating management, radiation monitoring supervision and supervision of service groups involved. The radiation monitoring group participates in the preparation of Radiation Procedures, but it is clearly understood that the responsibility for achieving a high degree of radiation safety rests with operating management.

4. Radiation Protection Instrumentation

Much effort has been directed toward the development of versatile, rugged and accurate instruments for use in the radiation protection program. The principal portable instruments used for beta-gamma dose rate monitoring are the CP and Juno. Geiger-Mueller (GM) instruments are used for beta-gamma contamination surveys. Alpha contamination surveys are made with several kinds of proportional counters. Some of the proportional counters have scintillation detectors and others have gas-filled ionization detectors. Portable instruments are distributed from a Battelle Northwest facility in the 300 Area where they are calibrated and serviced at intervals of one to four weeks depending on the type of instrument.

Instruments and instrument systems used in routine control activities such as remote area monitors, hand and shoe counters, air samplers, and semi-portable contamination detectors are assigned permanently to a Waste Management Facility. The routine performance checks and maintenance are carried out by personnel in the facility.

Hand and shoe counters are placed in several convenient locations and portable contamination survey instruments are available at the exits of many radiation zones. Dose rate monitoring instruments are also available where operating personnel can make effective use of them.

Fixed instruments for measuring gamma radiation intensities are installed in locations where there is a potential for abnormal radiation doses. Dose rates from these remote instruments are transmitted and recorded in centralized locations. Two types of alarms are provided for most of these fixed instruments. One is an "intermediate" alarm with a trip setting at a few hundred mrem/hour and the second trip point is at a dose rate level where radiation is an immediate hazard to personnel in the vicinity. The strip chart recorders are routinely checked by Radiation Monitoring personnel.

The concentration of radioactive material in building air is monitored by sampling and analyses. Fixed filter paper holders (sampling heads) on the building vacuum system are located in many work locations. Room air is drawn through filter paper at controlled rates. The filter papers are collected periodically, counted in the laboratory and the air concentration of radioactive material computed.

5. Radioactive Contamination of Air and Water

The concentrations of radionuclides in air and water concern internal exposure, and a list of maximum permissible concentrations provides a guide in establishing controls to achieve the occupational protection program objective of keeping internal exposure at insignificant levels. The tables below list the radionuclides encountered in Waste Management processes. Table XVII-1 A lists the maximum permissible concentrations in air and water for radiation workers (MPC for a 40-hour week).

The plant drinking water is routinely sampled and analyzed, and concentrations of radionuclides are found to be insignificant compared to the maximum permissible concentrations in water as shown in Table XVII-1 B. Where the potential air concentration of any radionuclide approaches the values shown in Table XVII-1, column 1, a radiation zone must be established and respiratory protection becomes a requirement for occupants.

These tables are displayed here to indicate the basis for the controls used in keeping the internal exposure of radiation workers at insignificant levels and to show the relative hazard of the different radionuclides encountered. Furthermore, the concentrations applicable to occupationally involved personnel as shown in A.can be readily compared to the permissible concentrations in B that apply to the non-occupational population. Further reference to the values in B are made under the discussion of the Environmental Protection Program.

6. Radiation Occurrences

The radiation protection program is based on control, and each time control is lost, an investigation is required. The established criteria for classifying the loss of control conditions are shown below.

6.1 Type A

- 1) A single radiation exposure to an employee exceeding 25 rems whole body, 150 rems skin of the whole body, or 375 rems hands, forearms, feet or ankles, or the summation of exposure during a calendar year exceeding 25 rems whole body.
- 2) A release of radioactive material off-site may result in any member of the general population receiving an exposure exceeding limits stated in AEC Manual Chapter 0524.

TABLE XVII-1

MAXIMUM PERMISSIBLE CONCENTRATION OF SELECTED RADIONUCLIDES IN AIR AND WATER (From RL-AEC Manual Chapter 0524 Appendix)

			Table			
Element (Atomic Number)	Isotope		Air (mc/ml)	Water (mc/ml)	Air (mc/ml)	Water (mc/ml)
Americium (95)	Am-241	s I	6x10-12 1x10-10	1x10 ⁻⁴ 8x10 ⁻⁴	2x10 ⁻¹³ 4x10 ⁻¹²	4x10 ⁻⁶ 2x10 ⁻⁵
	Am-243	s I	6x10-12 1x10-10	1x10 ⁻⁴ 8x10 ⁻⁴	2x10 ⁻¹³ 4x10 ⁻¹²	4x10-6 3x10-5
Barium (56)	Ba-140	s I	1x10-7 4x10-8	8x10 ⁻⁴ 7x10 ⁻⁴	4x10 ⁻⁹ 1x10 ⁻⁹	3x10 ⁻⁵ 2x10 ⁻⁵
Cerium (58)	Ce-141	S	4x10-7 2x10-7	3 x1 0-3 3 x 10-3	2x10 ⁻⁸ 5x10 ⁻⁹	9x10 ⁻⁵ 9x10 ⁻⁵
	Ce-144	S I	1x10 ⁻⁸ 6x10 ⁻⁹	3x10 ⁻⁴ 3x10 ⁻⁴	3x10-10 2x10-10	1x10 ⁻⁵ 1x10 ⁻⁵
Cesium (55)	Cs-137	S	6x10-8 1x10-8	4x10 ⁻⁴ 1x10-3	2x10-9 5x10-10	2x10-5 4x10-5
Curium (96)	Cm-242	s I	1x10 ⁻¹⁰ 2x10 ⁻¹⁰	7x10 ⁻⁴ 7x10 ⁻⁴	4x10-12 6x10-12	2x10 ⁻⁵ 3x10 ⁻⁵
	Cm-244	S I	9 x 10-12 1 x 10-10	2x10 ⁻⁴ 8x10 ⁻⁴	3x10-13 3x10-12	7x10-6 3x10-5
Iodine (53)	I -1 31	s I	9x10-9 3x10-7	6x10-5 2x10-3	lx10 ⁻¹⁰ lx10 ⁻⁸	3x10-7 6x10-5
Lanthanum (57)	Ia-140	s I	2x10-7 1x10-7	7x10 ⁻⁴ 7x10 ⁻⁴	5 x1 0-9 4 x1 0-9	2x10-5 2x10-5
Neodymium (60)	Nd-147	s I	4x10-7 2x10-7	2x10 ⁻³	lx10 ⁻⁸ 8x10 ⁻⁹	6x10-5 6x10-5
Niobium (41)	nd- 95	s I	5x10 ⁻⁷ 1x10 ⁻⁷	3x10-3 3x10-3	2x10 ⁻⁸ 3x10 ⁻⁹	1x10 ⁻¹ 4 1x10 ⁻¹ 4
Plutonium (94)	Pu-238	s I	2x10 ⁻¹² 3x10 ⁻¹¹	1x10 ⁻⁴ 8x10 ⁻⁴	7x10 ⁻¹⁴ 1x10 ⁻¹²	5x10-6 3x10-5
	Pu-239	S I	2x10-11	0x10 ⁻⁴	6x10 ⁻¹⁴ 1x10 ⁻¹²	5x10-6 3x10-5
	Pu-240	S I	2x10-12 4x10-11	1x10 ⁻⁴ 8x10 ⁻⁴	6x10-14 1x10-12	5x10-5 3x10-5
	Pu-241	S I	9x10-11 4x10-8	7x10-3 4x10-2	3x10-12 1x10-9	2x10 ⁻⁴ 1x10 ⁻³
	Pu-242	S I	2x10-12 4x10-11	1x10 ⁻⁴ 9x10 ⁻⁴	6x10-14 1x10-12	5 x 10 - 6 3 x 10 - 5

TABLE XVII-1, Continued

			Table		Table	В
Element (Atomic Number)	Isotope		Air (mc/ml)	Water (mc/ml)	Air (mc/ml)	Water (mc/ml)
Praseodymium (59)	Pr-143	S I	3x10-7 2x10-7	lx10 ⁻³ lx10 ⁻³	1x10 ⁻⁸ 6x10 ⁻⁹	5x10 ⁻⁵ 5x10 ⁻⁵
Promethium (61)	Pm-147	S I	6x10 ⁻⁸ 1x10 ⁻⁷	6x10 ⁻³ 8x10 ⁻³	2x10 ⁻⁹ 3x10 ⁻⁹	2x10 ⁻⁴ 2x10 ⁻⁴
Rhodium (45)	Rh-103m	S I	8x10 ⁻⁵ 6x10 ⁻⁵	4x10 ⁻¹ 3x10 ⁻¹	3x10-6 2x10-6	1x10 ⁻² 1x10 ⁻²
	Rh -105	S I	8x10-7 5x10-7	4x10 ⁻³ 3x10 ⁻³	3x10 ⁻⁸ 2x10 ⁻⁸	1x10 ⁻⁴ 1x10 ⁻⁴
Ruthenium (44)	Ru-103	S I	5x10 ⁻⁷ 8x10 ⁻⁸	2x10 ⁻³ 2x10 ⁻³	2 x1 0 - 8 3 x1 0 - 9	8x10 ⁻⁵ 8x10 ⁻⁵
	Ru-106	S I	8x10-8 6x10-9	4x10 ⁻⁴ 3x10 ⁻⁴	3x10 ⁻⁹ 2x10 ⁻¹⁰	1x10 ⁻⁵ 1x10 ⁻⁵
Strontium (38)	sr - 89	s I	3x10 ⁻⁸ 4x10 ⁻⁸	3x10 ⁻⁴ 8x10 ⁻⁴	3x10 ⁻¹⁰ 1x10 ⁻⁹	3x10-6 3x10-5
	Sr-90	S I	3x10 ⁻¹⁰ 5x10 ⁻⁹	4x10 ⁻⁶ 1x10 ⁻³	3x10 ⁻¹¹ 2x10 ⁻¹⁰	3x10 ⁻⁷ 4x10 ⁻⁵
Tellurium (52)	Te-129	S I	5x10-6 4x10-6	2x10 ⁻² 2x10 ⁻²	2x10-7 1x10-7	8x10 ⁻⁴ 8x10 ⁻⁴
Uranium	U - 235	S I	5x10 ⁻¹⁰ 1x10 ⁻¹⁰	8x10 ⁻⁴ 8x10 ⁻⁴	2x10 ⁻¹¹ 4x10 ⁻⁴	3x10 ⁻⁵ 3x10 ⁻⁵
	U - 238	s I	7x10-11 1x10-10	1x10 ⁻³	3x10 ⁻¹² 5x10 ⁻¹²	4x10-5 4x10-5
	U natural	S I	7x10-11 6x10-11	5x10 ⁻⁴ 5x10 ⁻⁴	3x10 ⁻¹² 2x10 ⁻¹²	2x10 ⁻⁵ 2x10 ⁻⁵
Xenon (54)	Xe-133 Xe-135		1x10 ⁻⁵ 4x10 ⁻⁵		3x10 ⁻⁷	
Yttrium (39)	Y-90	S I	1x10-7 1x10-7	6x10 ⁻⁴	4x10-9 3x10-9	2x10 ⁻⁵ 2x10 ⁻⁵
	Y-91m	S I	2x10 ⁻⁵ 2x10 ⁻⁵	lx10 ⁻¹ lx10 ⁻¹	8x10 ⁻⁷ 6x10 ⁻⁷	3x10 ⁻³ 3x10 ⁻³
	Y-91	S I	4x10-8 3x10-8	8x10 ⁻⁴ 8x10 ⁻⁴	1x10-9 1x10-9	3x10 ⁻⁵ 3x10 ⁻⁵
Zirconium (40)	Zr - 95	S I	1x10-7 3x10-8	2x10 ⁻³ 2x10 ⁻³	4x10-9 1x10-9	6x10 ⁻⁵ 6x10 ⁻⁵

- 3) An unplanned release of radioactive material which exceeds, in 24 hours, 5000 times the continuous non-occupational MPC $_{\rm a}$ or MPC $_{\rm w}$ shown in AEC Manual Chapter 0526 at the point of release.
- 4) An accident or radiation exposure which gives rise to an inquiry by members of the public or press or which is believed to have public information significance.

Type A radiation occurrences are investigated by a committee established by the Richland Office of the AEC (RL-AEC) and formal reports are issued.

6.2 <u>Type B</u>

- 1) A radiation exposure in a calendar quarter exceeding 3 rems whole body, 10 rems skin of the whole body, 25 rems hands, forearms, feet or ankles.
- 2) An employee's accumulated whole body exposure exceeds 5(N-18) rems, where N is the age in years.
- 3) An internal deposition of radioactive material where, on the basis of a preliminary internal dosimetry measurement, the exposure received over a period of one year will exceed the annual occupational exposure limits.

Type B radiation occurrences are usually investigated by a committee established by the manager of the facility involved. The Richland office of the AEC must approve of the committeemen selected. A formal report is also issued.

6.3 Type C

- Conditions existing where exposure meeting the criteria in A or B above were narrowly averted.
- 2) Internal deposition of radioactive materials where the estimated exposure received over the period of a year will exceed five percent of the annual occupational exposure limits.
- 3) An unplanned single exposure exceeding 1 rem to the whole body, 6 rems to the skin of the whole body, or 15 rems to the hands, forearms, feet or ankles.
- 4) Measurable deposition occurred in five or more persons but the limit set forth in item 2) above was not exceeded.

- 5) Radioactive materials may have been released to the atmosphere or to open roads at a rate exceeding 100 times established release controls.
- 6) A formal investigation report is considered necessary by the Personnel Protection Operation to provide an adequate radiation protection record.

A Type C occurrence is investigated by a committee chosen by the manager of the facility involved and a report compiled on a pre-printed form. This report includes an orderly presentation of the pertinent facts of the occurrence, a statement of conclusions of the investigators as to causes and a list of actions to be taken to minimize consequences and to prevent recurrence.

C. ENVIRONMENTAL PROTECTION

The purpose of the Hanford Environmental Protection Program is to prevent significant radiation exposure of people off-site as a consequence of plant operations. This purpose is accomplished by controlling the off-plant release of radioactive materials. In the operation of the Waste Management facility it is necessary to conform to the interest of those AEC Manual Chapters pertaining to the protection of individuals and population groups in uncontrolled areas. The maximum permissible dose for both external and external exposure to people off-site is stated in RL-AEC Manual Chapter 0524 Appendix II and is shown below.

	Dose (rem/year)	
Type of Exposure		Based on the average exposure of a suitable sample of the exposed population
Whole body, or gonads	0.5	0.17
Thyroid, bone, or GI tract	<u>,</u> 1.5	0.5

In addition to limits for total radiation exposure, AEC Manual Chapter 0524 also sets limits for radioactive isotopic concentrations in air and water (see Table XVII-1B). The limits for total radiation exposure and for radioactivity concentration in air and water are applicable to the operation of the total AEC facilities at Hanford. Radioactive Waste Disposal Guides, RL AEC Manual Chapter 0510 and RL Manual Appendix 0510, were developed to govern the waste disposal practices for the operation of single or multiple facilities at Hanford within these established limits. The major portion of the Environmental Protection Program is concerned with operating within the requirements of the Waste Disposal

Guides. Serious loss of off-site radiation exposure control would most likely occur with the uncontrolled release of radioactive liquid and gaseous effluents from the operation of the Waste Management facilities.

1. Radioactive Liquid Wastes

This section summarizes key features of the Radioactive Waste Disposal Guides and their impact on Waste Management Operations. Details of waste disposal controls are presented in references 25 and 27.

- 1) "Radioactive liquid wastes discharged into or onto the ground should be limited to the lowest practical amount". This guide is directly related to objective number four of the Waste Management Program, "Reduce to a minimum the amount of radioactive waste dispersed to the environs".
- 2) "The capability of quantitatively measuring actual releases of radioactive liquid wastes to the ground should be maintained". "Representative measurements should be made and documented for all radioactive liquid wastes discharged to ground, along with identification of the radionuclides so disposed". Operation within these guides is assured by sampling, analysis and volume measurement of the streams discharged to the ground. Samples are analyzed for total radioactivity as well as for long-lived radioisotopes of concern (e.g., Sr-90, Cs-137).
- 3) "Continuous instrument monitoring should be provided for all effluent streams having the capability of exceeding the release guides. An alarm should be used in conjunction with instrument monitoring to signal release rates in excess of operating controls". Following this guide has become an important environmental safeguard. Alternate disposal methods or retention procedures can often be initiated promptly upon detection of abnormal levels of radioactive materials in liquid effluent streams.
- "Each waste stream should be evaluated to determine compatibility with the proposed disposal facility and to assure adequate fission product retention. The effect of combining wastes should be evaluated before using a common disposal facility. Whenever practical, low activity wastes should not be discharged with higher activity wastes". "Characteristics of wastes, affecting the use-continuity of the disposal site, including radioactive and non-

radioactive materials, physical and chemical composition, volume flow rate should be defined. Permissible variations of each characteristic of the waste should also be defined". Special consideration should be given to liquid wastes which are immiscible with water". These guides have several obvious environmental protection points but in addition provide bases for practical methods of administering disposals.

- The number of subsurface liquid disposal sites for materials with biological significance and long half-lives should be kept to a minimum and should be identified. Liquid wastes exceeding 100 uCi/ml (high level), of relatively long lived radionuclides, should be contained and not discharged into the ground". The disposal in subsurface facilities is restricted, in practice, to those effluent streams containing less than 100 microcuries of spectrum activity or 2 microcuries of strontium-90 per milliliter of wastes. In this context, "spectrum activity" signifies fission products having concentration ratios approximating those present in Hanford fuels three to twelve months after reactor discharge.
- 6) "Liquid wastes discharged to large open ponds should be essentially free of radioactive materials. Concentrations of 5 x 10⁻⁵ uCi/ml (low level) should not be routinely exceeded". "Other than cooling water, disposal of liquid radioactive wastes should be subsurface facilities only". Sending radioactive liquids to open pools or ponds can lead to serious contamination spreads by wind borne residues from evaporation and/or filtration. The problem becomes complicated by pond-edge vegetation concentrating radioactive materials and by subsequent feeding of animals, particularly water fowl, on this vegetation. Even in the design of new or modified cooling water effluent lines, consideration is given to subsurface disposal, since experience has shown these streams become contaminated even when the probability appears to be very low.
- 7) "Liquids should not be discharged to facilities intended for the disposal of radioactive solid waste". The leaching of the solid waste by the liquids could mean loss of control of radioactive materials entering ground water.
- 8) "Surveillance over the movement of radioactive materials from underground disposal facilities should be maintained by monitoring the ground water from adjacent wells. Additional control measures, or termination of disposal should be effected when analysis of well water shows that radio-

nuclides such as cobalt-60, cesium-137 and strontium-90 intended for long-term retention by the soil are approaching one-tenth of their maximum permissible concentration as indicated in Table XVII-1 B. This surveillance is carried out primarily by Battelle Northwest Laboratory personnel who advise on the additional control measures to be taken.

2. Gaseous Radioactive Wastes

The Radioactive Waste Disposal Guides include the following definition: "Gaseous radioactive wastes are radioactive particulates, mists, and/or gases, contained in effluents released to the environment", (atmosphere). Key guides on these wastes are presented with comments on their application in Waste Management operations.

- 1) "Radioactive wastes released to the atmosphere should be minimized to the fullest extent technically and economically feasible". Here again is guidance on making efforts to achieve minimum releases rather than only striving to remain under prescribed levels of environmental pollution.
- 2) "Gaseous effluents should be filtered or otherwise controlled so on-site concentrations do not exceed the Standard in Table XVII-1 B." A double filtration system is maintained in Waste Management facilities that adequately controls the atmospheric concentrations under nearly all operating conditions.
- 3) 'The guide for total release of iodine-131 from all 200 Area facilities is 500 curies per calendar quarter". The guides for effluent release from each stack in the 200 Areas are:

	*Release	Guide (Ci/week)
Radionuclide	200-ft. Stack	Short Stack or Vent
Strontium-90	1	1 x 10 ⁻¹
Iodine-131	30	3
Plutonium (total)	1 x 10 ⁻²	1×10^{-3}
Uranium (natural)	4×10^{-2}	4×10^{-3}
Total of others (except tritium, carbon-14 and nobl		
gases)	20	2

*The policy is to strive to keep routine release at or below 10 percent of these values.

The ARHCO release limits are ten percent of the listed values and prompt corrective measures become necessary whenever data indicate ten percent value is likely to be exceeded. For design of effluent disposal equipment, the ten percent values are considered as the maximum permissible emission rates.

- 4) "Additional release guides should be developed when and where other radionuclides become present in concentrations that release might contribute significant exposure to individuals in uncontrolled areas". This guide would become pertinent upon consideration of handling different radioactive wastes than those under consideration at the time of this writing.
- 5) "Continuous instrument monitoring should be provided for gaseous effluent streams having a singificant probability of exceeding the release guide rates of emission. An alarm should be used in conjunction with instrument monitoring to signal releases in excess of operational controls". This type of instrumentation is in place and operating. Corrective measures need to be taken upon sounding of the alarm.
- 6) "Continuous sampling capability should be provided for gaseous effluent streams having a significant probability of exceeding one percent of a release guide value".

 "The capability of quantitatively measuring any radionuclide having a significant probability of being in a gaseous effluent in concentrations of one percent of release guide value or in concentrations that release might contribute significant exposure to individuals in uncontrolled areas". The sampling and analytical program meets the intent of these guides and reports the quantity of radioactive materials released to the atmosphere from the disposal of gaseous wastes to the RL AEC.

3. Other Aspects of Contamination Control

The most likely ways losses of off-plant radiation exposure control would be sustained are through releases of radioactive liquid or gaseous wastes. However, there are other ways control might be lost that are worthy of brief review.

3.1 Solid Waste

The disposal of solid radioactive wastes by burial has little direct relationship to possible significant radiation exposure of people off-site, since contamination control precautions taken for protection of the plant site and personnel adequately prevent loss

of control off-site. However, due to the quantity of radioactive material deposited in 200 Area burial grounds and cribs, control measures will be necessary at the disposal locations of both solid and liquid waste for many generations. These control measures include limitations on access and on use of the land. The integrity of a terminated disposal site needs to be maintained to provide control of vegetation and animals, soil stabilization, adequate backfill, and visibility of site boundary markers. Portions of the Radioactive Waste Disposal Guides (RL AEC Manual Chapter 0510) pertain directly to solid wastes and the traditional solid waste disposal methods follow the guidance provided.

3.2 Containers, Vehicles, and Personnel

The loss of control of radioactive materials from Waste Management facilities could result from undetected contamination on the surfaces of containers, vehicles, and personnel transported off-site. In general, the occupational protection procedures supply adequate directions for contamination control within the plant and prevent transport of contamination off-site. Guidance on the packaging, shipping, receiving, and transferring of radioactive material is given in RL AEC Manual Chapter 0550, Part III, Chapter 2 Appendix. This guidance is based primarily on regulations promulgated by the Interstate Commerce Commission (ICC), Title 49, Code of Federal Regulations, Parts 71-78. The ICC regulations form the basis for all other domestic regulations concerning radioactive materials in transport. Radiation procedures at individual facilities and provisions of the ARHCO Manual of Radiation Protection Standards and Controls follow the guidance of RL AEC Manual Chapter 0550 for intercontractor shipments and off-site shipments as well as providing rules for shipment between ARHCO facilities.

3.3 Explosion, Fire, Enemy Action, Earthquake

Explosion, fire, enemy action, earthquake, etc could result in the loss of control of radioactive materials from Waste Management facilities. Although radiation exposures to personnel resulting from such catastrophic events are not regulated by the same limits as radiation exposure from operational events, effort needs to be made to minimize the probability of occurrence and/or reduce the severity of the potential consequences. Safeguards were taken in the design of Waste Management facilities that were specifically directed toward prevention of serious spreads of contamination even from these events. Furthermore, operating procedures need periodic review to assure adequate off-site radiation control under any circumstance.

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PART VII, Continued

SAFETY

CHAPTER XVIII

DECONTAMINATION OF SURFACES

 $\mathbf{B}\mathbf{y}$

N. P. Nisick

Personnel Protection Operation

PART VII: SAFETY, Cont.

CHAPTER XVIII

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CHAPTER XVIII

DECONTAMINATION OF SURFACES

Decontamination is employed at B-Plant for the removal of all or part of the residual radioactive contamination from operating equipment and areas to improve working time limits and to control the spread of contamination. This chapter presents a summary of information concerning decontamination methods. The ability to decontaminate a surface is determined by the material the surface is made of, protective coatings (if any), chemical composition of the contaminant, the length of time the surface has been contaminated, and the amount of physical effort used in decontamination.

A. GENERAL

1. Introduction

The need requiring the removal of radioactive contamination from equipment or areas varies widely. The amounts and concentrations of radioactivity within B-Plant cover a wide range and include some of the highest concentrations on site. The bulk of the radioactivity entering the plant is acid fission product salt waste originally generated at the Purex Plant. This fission product waste will pass successively through feed treatment, solvent extraction, and concentrating vessels with the more noxious fractions going to in-canyon tank storage and the less noxious but more voluminous fractions going to ITS (in-tank solidification) at one of the 200-E Area tank farms.

Because of the nearly universal use of stainless steel in the construction of all B-Plant process equipment, decontamination of process equipment is largely a matter of removing contamination from stainless steel. This particular material has received considerable study, and considerable success has been realized. The structural and support components of the plant are fabricated almost entirely of mild steel or reinforced concrete. These materials if not covered with a protective coating are extremely difficult, if not impossible, to decontaminate. In the processing areas, all non-stainless steel components have been painted. This permits the use of reagents for decontamination that might otherwise react with the unprotected surface.

Decontamination techniques developed to date have proven highly successful for many specific needs, but may not be the optimum techniques for all new B-Plant application. The following sections describe reagents and techniques which have been used successfully for decontamination. Continued experience and study of decontamination should lead to even better reagents and techniques.

2. Terminology

The procedure of decontamination has given rise to the use of some special terminology. The term "decontamination" is used to designate the removal of radioactive material from solid surfaces.

"Contamination" or "contaminants" are the radioactive materials present on the solid surface. Decontamination methods and techniques are the means utilized to remove the radioactive material. They may be generally classified as (a) physical--total or partial removal of the contaminate surface; (b) chemical--reaction with the radioactive material or the larger mass of the associated non-radioactive materials to convert them to a soluble or extractable condition; and (c) physicochemical--suspension by surface-active agents.

3. Principles of Decontamination

A review of decontamination techniques reveals many conflicting and varied decontamination procedures. One can conclude that no "cut and dried" methods for decontamination exist and no reagent is universal for decontamination. In most cases the best decontamination agent for a specific problem must be developed empirically. However, the lack of exact knowledge does not call for a random approach to decontamination.

The mechanisms governing surface contamination and decontamination are not yet too well understood. However, laboratory tests and decontamination of process equipment indicate some of the principles of contamination-decontamination to be as follows:(3)

- a. The art of good decontamination requires the control of radioactivity at all times. Planning and the subsequent use of a well-thought-out decontamination procedure is helpful no matter how small the job.
- b. Surfaces contacted by "spills" should be flushed or scrubbed before the activity has had a chance to go to dryness. This applies also to the interior or process vessels.
- c. Removal of a thin film or layer of the contaminated surface is necessary for achievement of maximum decontamination in the shortest period of time.
- d. Complexing, chelating, and metathesizing agents are of little value after the activity has gone to dryness.
- e. Successive use of several reagents over short periods of time (one to two hours) appears to produce better decontamination than the use of a single reagent over an extended period of time (twelve to sixteen hours). It appears that one reagent may prepare a contaminated surface for attack or removal of radioactivity by a second reagent that follows.

- f. Good agitation (in many instances good old "elbow grease") is necessary for the successful use of any reagent. Stagnant or static surface films produce little or no decontamination.
- g. The efficiency of a given reagent usually increases as the temperature increases; however, the maximum practical temperature may be limited by corrosion or boiling.
- h. Dry-solids contaminants such as dusts should not ordinarily be placed into solution to expedite removal. Vacuum cleaning or flushing with a reagent that does not appreciably dissolve or convert dry solids into the ionic form is much more desirable.
- i. In general, surfaces may be protected from radioactive contaminants by reduction of the surface free energy (used here as analogous to ease of wetting). Coating exposed surfaces with relatively non-wetting membranes such as paint or non-polar plastics is always desirable.
- j. Strippable coatings or other inert membranes where such application is feasible presents the easiest and perhaps the most successful method for decontamination at this time.

4. Factors Influencing Decontamination

The following are some of the factors affecting decontamination procedures in B-Plant.

- a. The inactive anions and cations in the B-Plant process solutions do not normally form insoluble compounds. Hence, any deposits formed by exceeding the saturation point should be removable with water flushes.
- b. B-Plant incorporates design for confining process materials within the interior or processing equipment.
- c. The employment of some continuous-flow equipment in the B-Plant process necessitates some mechanical complexity with the associated possibility of occasional need for equipment replacement and adjustment.

5. Provisions for Application of Decontamination Procedures

5.1 Protective Coatings

Structural elements in the plant have been either protected with a chemically resistant coating such as "Amercoat" or constructed on stainless steel. Thus, all exposed concrete within the cells has been protected with "Amercoat". Structural steel elements and certain equipment fabri-

cated from carbon steel have been protected with "Amercoat". This type of construction facilitates removal of the gross radioactivity by simple water flushing.

5.2 Materials of Construction

Process equipment normally wetted by process solutions has been fabricated almost entirely from stainless steel. Some inportant but relatively small elements utilize alloys such as Stellite or fluorocarbon plastics. All of these materials have exceptionally good resistance to chemical attack and are readily decontaminable.

5.3 Design Features Facilitating Decontamination

All equipment normally in contact with process solutions is designed to be fully drainable and flushable. The process equipment also incorporates seals on all pump and agitator shafts, which, in conjunction with the vacuum vent system, effectively confine radioactive material to the interiors of process equipment.

B. STAINLESS STEEL

The process equipment components (tanks, lines, pumps, etc.) of the B-Plant are fabricated almost entirely from the 300 series stainless steels. The specific type employed has been chosen on the basis of corrosion resistance and the type of fabrication required. The decontamination of stainless steel is consequently of prime interest. Since the problem of decontamination is solved by an empirical approach, discrimination between the types of 300 series stainless steel is not justifiable.

The contamination of stainless steel can be placed in four categories:
(a) alpha contaminants on internal surfaces, (b) alpha contaminants on external surfaces, (c) beta and/or gamma contaminants on interior surfaces, and (d) beta and/or gamma contaminants on exterior surfaces. The recommended procedures for removal are based on empirical 200 Area results. In general, the least corrosive effective decontaminant should be used so that attack upon the equipment may be maintained at a minimum.

Internal surfaces should be first in the sequence of decontamination because of the greater freedom and control associated with this operation and because these surfaces contain large amounts of easily removably contamination. To produce results of the greatest value, decontamination flushes should be routed from areas of low radioactivity into areas of high radioactivity. Decontaminants that are combined with process streams for disposal must be compatible with the process solutions so that normal plant production is not interrupted. Cribbable decontaminating solutions must not contain reagents at the time of cribbing that have an adverse affect on the removal of radioactive material by the soil. (5)

1. Alpha Contamination

1.1 Internal

Alpha (notably U and Pu) contamination inside process vessels assumes relative little importance and only where the fission-product beta and gamma activites associated with it have reached comparatively low levels. Little U and Pu will normally be associated with the material being processed.

Nitric acid is the preferred decontamination reagent, as the chemistry of the principal alpha contaminants favors solution in this medium. Sixty percent acid is the most effective, although lesser concentrations have been used successfully. Normally a temperature of 25 to 50°C is adequate. Generally, physical and chemical principles indicate that higher temperatures would be more effective although there is very little practical decontamination experience to support this.

1.2 External

The problem of decontaminating external surfaces is complicated by the possibility of extending the contamination to otherwise clean areas. Therefore, caution must be exercised since the need for decontaminating surfaces adjacent to those originally contaminated may be increased by unrestricted and uncontrolled flushing.

The following precautions should be observed in decontaminating external surfaces:

- (a) The redistribution of existing contamination should be limited to the smallest possible area.
- (b) Normally the surface should not be flooded with water or other decontaminating agents.
- (c) The surrounding surfaces which might be wetted during the decontamination treatment should be protected.
- (d) Contaminated materials such as tools, rags, swabs, or gloves should not be placed in uncontaminated areas. Disposable containers such as bags or boxes should be used for storing all cleaning supplies.

The choice of decontamination reagents is based on factors such as the original surface finish, necessity for preserving the finish, and limitations of drainage facilities for solvents or highly radioactive chemical agents. The materials listed below have been found to be useful for room temperature application:

- (a) Nitric 15% to 57%
- (b) Oxalic 5% Nitric 3%
- (c) Caustic 25%
- (d) Permanganate-bisulfite. The sequence of operations consists of scrubbing the surface with saturated KMnO₁, solution, removing the KMnO₁, by wiping, scrubbing the surface with 10% NaHSO₃ solution, and flushing the surface with water.
- (e) Organic solvents; e.g. Stoddard Solvent, CCl4.
- (f) Abrasives; e.g. Bon Ami
- (g) Detergents; e.g. Tide
- (h) Proprietary reagents; Turco 4306-B, 4502, 4518

2. Beta and Gamma Contamination

2.1 Internal

Since most of the surfaces are contaminated with beta and/or gamma contaminants, it is general practice to use nitric acid for the first flush. Although beta and/or gamma contaminants are not completely removed by nitric acid, it is believed that it is useful in removing the major portion of non-radioactive materials constituting the film in which the radioactivity is bound.

Oxidizing, reducing, and complexing agents have been used successfully. A solution of sodium tartrate in sodium hydroxide has been found to be effective. The concentrations of sodium hydroxide and sodium tartrate are not well defined, but 50 to 20 percent NaOH and 1.25 to 10 percent Na₂C₄H₄O₆ have been recommended. The addition of hydrogen peroxide to the alkaline tartrate improves its efficiency.(8) Oxalic acid, citric acid, and nitric acid give approximately equal decontamination results. HNO₃ is often preferred because it is the cheapest and most readily available agent.

The use of mixtures of nitric acid and sodium fluoride to remove the final contamination is often necessary. A mixture of 3 percent fluoride and 20 percent nitric used at room temperature has been found to be an effective reagent. It decontaminates rapidly without excessive corrosion of stainless steel equipment. The degree of corrosion and of decontamination is not necessarily related.(9) Although some surface corrosion may be necessary for the removal of tightly bound contaminants, corrosion is not always accompanied by good decontamination. At higher temperatures, Al(NO₃)3·9H₂O (ANN) is added for corrosion control.

Experience at Hanford has shown that a basic solution of permanganate (1.5% $\rm KMnO_4$, 0.5% NaOH) is very effective for the removal of fission product contamination, especially of radioruthenium.(2) The ruthenium is believed to be oxidized to a soluble ruthenate, probably $\rm RuO_2$ which is soluble in a basic solution.(6)

For successful internal decontamination, frequent changes of decontamination solutions are more beneficial than extended contact time. It has been found that the most rapid dissolution of radioactive contaminants in the decontamination solutions occurs in from 12 minutes to 2 hours, depending upon the solution used. (7)(8) Considerations governing the frequency of solution change include the cost of the disposal of the solution. Adequate agitation is a prime requisite of successful internal decontamination and elevated temperatures usually add to the effectiveness of the flushing solution.

In addition to these liquid decontaminants, live steam seems to assist materially in the decontaminating process. The mechanism of decontamination with steam is not fully understood. It is conjectured that it penetrates the minute recesses inaccessible to liquids. It is also believed that it acts by the exertion of some abrasive action.

2.2 External

The plant areas in which external beta and gamma contamination is of potential concern coincide with those in which beta-gamma internal contamination levels are high. This problem is the most difficult of the four discussed because (a) the flush solution is not confined by equipment, and (b) precautionary measures to restrict the spread of contamination by the decontamination agents cannot be employed nearly as effectively as with alpha contamination because the range of the gamma rays often prohibits the approach of personnel to the contaminated surfaces. The decontaminating agents recommended for the removal of internal contamination (under B 2.1 above) can also be employed for the removal of external contamination. To assist in carrying the contamination from the decontaminated areas, sump, or drain, the entire area undergoing decontamination should be liberally flushed with water as a concluding treatment.

The method of applying external decontaminating agents will vary widely with the amount of radioactivity present in the area to be decontaminated. If the radiation level is sufficiently low to permit entry of personnel, the solutions may be applied manually by mop or preferably by brush or broom. This application may be followed by steam cleaning and water flushing or water flushing alone.(2)

If the radiation level does not permit entry by personnel, the decontamination solutions may be applied from the closest point of vantage by means of a pump or jet and a spray nozzle. Repeated applications of the decon-

tamination solutions followed by water flushes or steam cleaning and water flushing usually reduces the radiation sufficiently to permit eventual entry by personnel.

3. Procedure for Decontamination

The general procedure for internal decontamination usually starts with a nitric acid flush to remove any process solutions present. Decontamination itself is then accomplished by successive flushes with suitable decontaminating agents kept in the vessels with agitation, generally at elevated temperatures for the proper period of time.

External decontamination is accomplished by application of a suitable reagent by the most effective means available. Abrasive action applied either manually or by some other means, such as high pressure water, usually assists in contamination removal. The contamination loosened from the surface should be flushed away with water.

In order to select a suitable decontamination reagent, a gamma scan of the contamination may be used to determine the specific fission products present.

C. OTHER MATERIALS OF CONSTRUCTION

1. Iron and Carbon Steel

Iron and carbon steel have been employed for machine and structural elements (e.g. electric motors, jumper braces) which are not normally wetted by process solutions. As installed, these elements are protected by a chemically resistant coating, "Amercoat". The type of contamination expected on these surfaces follows the pattern given in Section C of this chapter. Experience with decontamination of unprotected iron or steel has been generally disappointing. Complete removal of the contaminated piece usually is the most practical solution to the problem.

Nitric acid, although effective in decontaminating stainless steels, is not effective in the decontamination of unprotected iron and steel. The degree of readsorption of the highly diluted, radioactive elements by the freshly formed, unoxidized metallic surface appears to create a practical obstacle which limits the decontamination. In addition to the ineffectiveness of acidic solutions, their use increases the difficulty of successfully removing the contamination with subsequent decontaminating agents because of the surface-etching effect of the acid.

Alkaline solutions (sodium carbonate, alkaline permanganate) and detergents have been found the most effective of the agents tried.(2)

2. Concrete

Concrete, in the absence of a protective surface coating, retains contaminants to a high degree. Chemical or physicochemical reagents have not been effective in removing either chemically fixed or physically adsorbed radioactivity. B-Plant has been designed and constructed on the basis that concrete surfaces will always be protected by a chemically resistant coating (Amercoat). In the event of contamination which penetrates through the concrete base material, only two courses appear practical: (a) physical removal of the contaminated surface, or (b) shielding of the contaminated concrete surface to permit necessary maintenance work in the immediate area of contamination.

Experience has indicated that spots of gross contamination imbedded in unprotected concrete may be reduced somewhat by repeated alternate treatments with sodium carbonate paste (or solution) and dilute (3 to 5 percent) nitric acid. The effervescing action produced by the two reagents helps to lift some of the contamination out of the concrete so that it can be flushed away with water. However, because of the destructive action of the nitric acid on the concrete, part of the remaining contamination is usually forced deeper into the concrete, thus making complete removal of the contamination practically impossible.

3. Glass

Glass finds its greatest application in the construction of laboratory equipment. Accordingly, the type of contamination may vary widely. A five percent solution of NHLHF2 has been effective in decontaminating glass at ORNL. Etching does not result if the time of contact is kept to a minimum (a few minutes).(4)

Nitric acid in concentrations of 20 to 60 percent is also used effectively for decontaminating laboratory glassware. The more concentrated solutions are used for soaking the equipment while the less concentrated are usually used for swabbing. Laboratory cleaning solution (95 percent sulfuric acid saturated with sodium dichromate) is another effective agent for decontaminating glassware.

4. Wood

Wood is utilized primarily for structural or equipment items, such as office fittings and furniture, not normally subject to contamination. Occasional temporary requirements may result in the introduction of wood into process or laboratory areas. Decontamination of wood is extremely difficult because of its porous, absorbent structure. (4) Protection from fixation of activity is dependent upon the applied protective coating. In the event that contamination penetrates the protective coating, either partial or complete removal of the wood appears to be the most practical and economical procedure.

D. PROTECTIVE COATINGS

Protective coatings employed in B-Plant for application to steel and concrete surfaces are Amercoat No. 33, 55, and 74. These coatings provide a protective film that is smooth, chemically resistant to process and decontaminating solutions, and not readily wetted by them.

Since the protective films are the only barrier to essentially permanent contamination of the base materials, it is important to use a decontaminant which will inflict the minimum damage to the coating. Highly reactive materials such as nitric acid or caustic soda should be used as sparingly as possible and only in dilute solutions. The following agents, listed in order of preference, are recommended:

- a. Water
- b. Detergent solutions
- c. Complexing agents, oxalates, citrates, tartrates
- d. Alkaline permangarate-oxalic acid. This requires direct manual application of 1-1/2 percent KMnO₄-1/2 percent NaOH solution for a 5 to 10 minute period followed by an application of 1 to 2 percent NaOH solution and, in turn, by a water flush. Manual application of 5 percent oxalic acid followed by a water flush completes the procedure.

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